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Title page

Title: Critical review of magnetic biosorbents: Their preparation, application, and regeneration for wastewater treatment

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Critical review of magnetic biosorbents: their preparation, application, and regeneration for wastewater treatment

Abstract: The utilisation of magnetic biosorbents (metal or metal nanoparticles impregnated onto biosorbents) has attracted increasing research attention due to their manipulable active sites, specific surface area, pore volume, pore size distribution, easy separation and reusability that are suitable for remediation of heavy metal(loid)s and organic contaminants. The properties of magnetic biosorbents (MB) depend on the raw biomass, properties of metal nanoparticles, modification/synthesis methods and process parameters which influence the performance of removal efficiency of organic and inorganic contaminants. There is a lack of information regarding the development of tailored materials for particular contaminants and the influence of specific characteristics. This review focuses on the synthesis/modification methods, application and recycling of magnetic biosorbents. In particular, the mechanisms and the effect of sorbents properties on the adsorption capacity. Ion exchanges, electrostatic interaction, precipitation, and complexation are the dominant sorption mechanisms for ionic contaminants whereas hydrophobic interaction, interparticle diffusion, partition and hydrogen bonding are the dominant adsorption mechanisms for removal of organic contaminants by magnetic biosorbents. In generally, low pyrolysis temperatures are suitable for ionic contaminants separation, whereas high pyrolysis temperatures are suitable for organic contaminants removal. Additionally, magnetic properties of the biosorbents are positively correlated with the pyrolysis temperatures. Metal-based functional groups of MB can contribute to an ion exchange reaction which influences the adsorption capacity of ionic contaminants and catalytic degradation of non-persistent organic contaminants. Metal modified biosorbents can enhance adsorption capacity of anionic contaminants significantly as metal nanoparticles are not occupying positively charged active sites of the biosorbents. Magnetic biosorbents are promising adsorbents in comparison with other adsorbents

including commercially available activated carbon, and thermally and chemically modified biochar in terms of their removal capacity, rapid and easy magnetic separation which allow multiple reuse to minimize remediation cost of organic and inorganic contaminants from wastewater.

Keywords: Adsorption; Heavy metal(loid)s; Organic contaminants; Metal nanoparticles; Biochars; Wastewater treatment.

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1. Introduction

Contamination of water and its remediation has emerged as a global and escalating problem (Shannon et al., 2009). A particular concern is the presence of heavy metal(loid)s, e.g. cadmium and arsenic, in increasingly elevated concentrations in streams, lakes, and rivers. This leads to bioaccumulation and successive biomagnification of heavy metal(loid)s in living organisms, including plants, animals, and human beings (Nordstrom, 2002; Smedley and Kinniburgh, 2002; Weber and Morris, 1962). Heavy metal(loid)s contamination of water has been observed all over the world, in particular in countries like Turkey, China, India, and Bangladesh where the heavy metal(loid)s concentrations in surface water exceed permissible limits in many regions due to natural processes and human activities (Azhar et al., 2015; Li and Zhang, 2010; Liao et al., 2017; Michael, 2013; Naidu et al., 2006; Nestle and Kimmich, 1996; Paul, 2017; Reza and Singh, 2010; van Geen et al., 2003; Varol and Şen, 2012). Similarly, organic contaminants have negative impacts on public health and aquatic environments, even at low concentrations. Priority emerging contaminants are chemicals released from antibiotics, pesticides, pharmaceuticals, and personal care products (PCP) which are discharged into the aquatic environments without adequate remediation. However, there is still inadequate information on the remediation technology, environmental fate and toxicological impacts of the emerging contaminants (Dong et al., 2015; Naidu et al., 2016a; Naidu et al., 2016b). For sustainable water management and protection of human and environmental health, the toxic inorganic and organic contaminants should be efficiently removed from wastewater before being discharged or further used.

Many strategies have been developed for wastewater treatment, including chemical precipitation, ion exchange, ultrafiltration, reverse osmosis, electrodialysis, coagulation, flotation, and adsorption (Ali and Gupta, 2007). Of these methods, adsorption has been recognised as the most promising technology for wastewater treatment (Ali and Gupta, 2007; Cledon et al., 2018; Ge and Li, 2018a). Adsorption technology is easy to operate, cost-efficient, and environmentally friendly in comparison with other remediation technologies (Ali and Gupta, 2007). It can adsorb a wide range of contaminants including organic, inorganic, and biological substances (microorganisms) covering both soluble and insoluble compounds (Ali and Gupta, 2007; de Andrade et al., 2018; Duan et al., 2018; Ge and Li, 2018b; Lim et al., 2009; Liu et al., 2015a; Zhang et al., 2017a). Adsorption technology can be applied to clean potable water, wastewater, industrial water, and water for other uses. However, there is a lack of suitable adsorbents with sufficient adsorption capacity, easy separation and regeneration for further uses.

Metal induced biosorbents can be promising options in compared to activated carbon, low-cost industrial waste materials, chemically modified plant waste, polymers, minerals, metal nanoparticles due to high removal performance, and magnetic properties (Charpentier et al., 2016; Farrukh et al., 2013; Khajeh et al., 2013; Lim et al., 2009; Nisticò et al., 2017; Yan et al., 2014a; Yang et al., 2016; Zhu et al., 2014b). However, biosorbents showed limited efficiency in removal of contaminants at trace levels, whereas metal nanoparticles perform efficiently (Ali, 2012). The blockage of active sites in metal nanoparticles by natural organic matter and other suspended particles limit its direct application for real wastewater treatment. Additionally, metal nanoparticles tend to aggregate due to weak van-der-Waals forces and consequently challenges its application for remediation purpose. To improve the dispersion and usability of the metal particles, it could be modified with carrier materials (e.g. carbonaceous materials) to improve their usability for wastewater treatment (Zou et al., 2016). Thus, a sustainable carrier materials is essential to tackle these issues. Metal induced biosorbents are generally produced by modifications of biomass or biochar (pyrolysis product of biomass) with metal or metal nanoparticles including iron, nickel, titanium, zirconium, zinc, copper and alloys (Chen et al., 2011; Lunge et al., 2014; Mohan et al., 2014a; Nistico et al., 2018; Reddy and Lee, 2014; Wang et al., 2018a; Wang et al., 2015e; Wang et al., 2015f; Wang et al., 2015g; Wang et al., 2018b). Thus, magnetic biosorbents have two major components: metal or metal oxide nanoparticles and biosorbents. Biosorbents are usually prepared via carbonization of low-cost agricultural, forestry and municipal green wastes including pinewood, sawdust, sugarcane bagasse, almond shells, olive bagasse, peanut shells, manures, grass, oak wood, and straw (Cledon et al., 2018; Fomina and Gadd, 2014; Hass and Lima, 2018; Liu et al., 2017a). Biochar is a carbon-rich material that is formed by pyrolyzing biomass in a limited oxygen environment to ensure stability of the materials in the wastewater system (Lehmann and Joseph, 2015). The properties of biochar largely depends on the pyrolysis temperature, feedstocks materials and process parameter which already reviewed critically (Kloss et al., 2012; Shen et al., 2019; Zhao et al., 2018). Metal or metal nanoparticles that are normally used include iron, copper, zirconium, titanium, zinc, cobalt nickel and alloys as a form of metal, nitrate, carbonate, sulphate and their oxides (e.g. magnetite (Fe_3O_4), $FeCl_3$, Fe_2NO_3, $Fe_2(SO_4)_3$, zero-valent iron (nZVI), and maghemite (Fe_2O_3, γ-Fe_2O_3)) (Dinari and Tabatabaeian, 2018; He et al., 2018; Liu et al., 2009; Mohammed et al., 2017; Rajapaksha et al., 2016; Zhou et al., 2018a; Zubrik et al., 2018). Magnetic biosorbents enhance the development of the specific surface area, pore size, and surface functionality to contribute higher adsorption capacity and facilitate recovery of biosorbents (Charpentier et al., 2016; Chen and Pan, 2013; Hayashi et al., 2010; Liu et al., 2015b; Tan et al., 2016b; Yin et al., 2017; Zhu et al., 2014b).

In recent years, many studies have reported the effectiveness of synthesised magnetic biosorbents for the removal of heavy metal(loid)s, dyes, pesticides, antibiotics and other emerging contaminants (e.g. Cd, Cr, Sb, As, Pb, Ba Hg, and PFAS) (González Vázquez et al., 2016; Hao et al., 2017; Huang et al., 2017; Huang and Keller, 2013; Jiang et al., 2018a; Karunanayake et al., 2016; Meng et al., 2017; Noor et al., 2017; Yan et al., 2015b; Yang et al., 2016; Zhao et al., 2015; Zhu et al., 2017). The adsorption capacity largely depends on feedstock materials, their modification, developed functional groups and the types of contaminants (Nistico et al., 2018; Wang et al., 2015e; Zhang et al., 2013b). Moreover, magnetic biosorbents can be modified and tailored for removing particular target contaminants. Carbo-thermal reduction of transition-metal-functionalized porous carbon nanostructures received enormous scientific attention due to its low production cost, reusability for commercial application especially environmental and energy sector (Shen, 2015). There are several review articles on the magnetic adsorbents for wastewater treatment (Mehta et al., 2015; Sivashankar et al., 2014b; Thines et al., 2017c). For example, the carbo-thermal modifications of metal induced biochar composites were critically reviewed for remediation of oxyanion including AsO_4^{3-}, AsO_3^{3-}, CrO_4^{2-}, NO_3^{-} and PO_4^{3-} (Li et al., 2018), including the effects of metal dose and pyrolysis conditions on the surface properties, stability, and eco-toxicity of the metal induced materials (Li et al., 2018). The interaction mechanism of nZVI induced biochar with heavy metals, nitrates, and organic compounds in soil and water system were reviewed highlighting on the synthesis condition of nZVI/BC in term of electrical conductivity, crystallite size and dispersion of nZVI on the biochar surface (Wang et al., 2019). The sludge-derived biochar from sewage sludge produced via carbonation process was reviewed for environmental remediation (Mian et al., 2019). However, limited information is available on different synthesis method and reuse of magnetic biosorbents for removal of organic and inorganic contaminants. While this review focuses on the different synthesis method for magnetic biosorbents, including the advantages and disadvantages, attempting to understand the sorption performance and mechanism in relation to the properties of the adsorbents. A systematic comparison of adsorption capacity for a wide range of adsorbents including minerals, polymers, activated carbon, biosorbents, nanomaterials, nanocomposite and magnetic biosorbents for Cd, As are also depicted. Regeneration, reuse, and recommendations for future research on magnetic biosorbents are also outlined. The information will be useful for understanding and application of modification methods for further development of magnetic biosorbents considering different contaminants removal from wastewaters.

2. Synthesis of magnetic biosorbents for wastewater treatment

The properties of magnetic biosorbents depend on the properties of their biomass, metal nanoparticles, modification methods and process parameter including temperature, duration of pyrolysis, and size of biosorbents (Alizadeh et al., 2018a; Dinari and Tabatabaeian, 2018; He et al., 2018; Reguyal et al., 2017a; Zhou et al., 2018a). Metal doped biosorbents improve yield of adsorbents, thermal stability, porosity, surface heterogentiy and crystalline structure (Chen et al., 2011; Reddy and Lee, 2014; Wang et al., 2015f). For example, a rough surface (heterogeneous) and abundance of porosity of magnetic biosorbent were evident from SEM micrograph and EDS image due to the presence of metal nanoparticles on the biochar surface, which are different from unmodified biochar (Figure SM1). The presence of crystal metal oxide is identified according to the XRD spectra (Figure SM1).

The properties of the biomass used for making magnetic biosorbents are critical in determining the properties of magnetic biosorbents. The elemental compositions of biosorbents vary significantly, and their changes as a result of the pyrolysis process and biomass types are discussed comprehensively by Liu et al. (Liu et al., 2017a). Variations in the major elemental compositions and degradation pathways of biomass materials result in diverse sorbent materials. For example, silicon-rich biosorbents including sugarcane bagasse, rice husk, and wheat husk can contribute to the improvement of adsorption capacity due to the abundance of the silicon-based functional group and structure (Li et al., 2019a; Xiao et al., 2014; Xu and Chen, 2015). Similarly, manure derived biochar contains nitrogen, potassium, phosphorus, calcium, magnesium, and iron that can also improve adsorption capacity due to the abundance of functional groups. While the biochar yield is reduced due to the presence of alkali metals which can produce more ash by oxidation of the alkali metals (Cabilovski et al., 2014; Cao and Harris, 2010; Cao et al., 2009; Nasir et al., 2014). Figure 1 illustrates the elemental compositions of carbon (C), hydrogen (H), oxygen (O), and nitrogen (N) of different biomass feedstock materials, biochars, and magnetic biosorbents. The variation of C content in biochar and magnetic biosorbents also depend on the pyrolysis conditions including reactor type, gas flow, time and most importantly pyrolysis temperatures (Sun et al., 2014; Weber and Quicker, 2018). For example, The H content is higher in raw biomass, and decreases in biochar and magnetically modified biosorbents (Figure 1). Similarly, the raw biomass contains around 41- 48% of O while it decreases after pyrolysis and magnetic modification (Figure 1). Conversely, N content increases in biochar comparing to biomass due to the vaporisation of H and O (Figure 1). The O content is influenced by the mixing ratio of biochar and metal oxide nanoparticles. The metal content of magnetic biosorbents generally varies from 1% to 20% based on the types and ratio of metal nanoparticles impregnated on the biosorbents (Baig et al., 2014a; Charpentier et al., 2016; González Vázquez et al., 2016; Hao et al., 2017; Lian et al., 2014; Lin et al., 2012; Meng et al., 2017; Noor et al., 2017; Sivashankar et al., 2014a; Thines et al., 2017c). The ratio of metal nanoparticles to biomass should be optimised as higher ratio may result in the nanoparticles covering most of the active sites of the biochar and significantly influencing adsorption capacity.

Cellulose, hemicellulose and lignin content as well as the pyrolysis state can also influence the development of surface functional groups (Cha et al., 2016; Liu et al., 2017b; Sharma et al., 2018; Zhang et al., 2017b) and chemical compositions (Fagbayigbo et al., 2017; Jing et al., 2014; Ramalingam et al., 2018; Zhang et al., 2013c) of the generated biosorbents. The cellulose, hemicellulose, and lignin percentages in biomass can vary from 20 – 60%, 20 – 40% and 20 – 45%, respectively (Yang et al., 2007). Hemicellulose decomposes at 470-540 ºC during pyrolysis whereas cellulose and lignin start breaking down at 500-600 ºC and 550 ºC -800 ºC, respectively (Lian and Xing, 2017; Yang et al., 2007). However, the temperature for decomposition may vary on the type of biosorbents and their chemical composition. The amount of lignin present in the raw biomass is the main contributor to biochar yield, whereas cellulose and hemicellulose generally introduce functional groups on the biosorbents surface, once it is chemically and/or thermally modified (Liu et al., 2015b; Yang et al., 2007; Yang et al., 2019c). Alkane and alkene bonds generally transfer into alcohol, aldehyde, and carboxylic acid as the temperature reaches 300-400 ºC while the hydroxyl, aldehyde groups, and a few aromatic structures will start forming at a temperature of 500-600 ºC. Over 600 ºC, the aromatic carbon structure and ash will start to form (Liu et al., 2015b). Functional groups develop on the biochar surface through the pyrolysis, oxidation, dehydration, and decarbonisation processes (Liu et al., 2015b). Alternatively, the reduction, hydration, and carbonisation processes can occur when reducing agents are present. Biosorbents modified with metal nanoparticles can be cracked and their C-O or COOH functional groups changes into C=O/C=C, which indicates that C-O-Fe acted as an electron acceptor for the reduction process (Zhu et al., 2017). Low pyrolysis temperature is effective for ionic contaminants removal due to abundance of O containing functional groups and developed surface charges whereas, high pyrolysis temperatures required to induce hydrophobic properties for organic contaminants removal. The variations in composition can result in the development of different functional groups, which are summarised in Table SM1. Metal or metal oxide doped adsorbents including nZVI (zero-valent iron), $CuFe_2O_4$, $ZnFe_2O_4$, Fe_3O_4, and Fe_2O_3 are widely used and can develop functional groups including Fe-O, Mn-O, and metal-O-metal-, which contribute to the adsorption capacity especially for ions (Guo et al., 2018; Liu et al., 2010; Senapati et al., 2011; Theydan and Ahmed, 2012). Magnetic biosorbents are also susceptible to further modification to tailor their sorption characteristics.

The ferromagnetic properties of magnetic biosorbents enable easy separation using a magnetic field after solid phase extraction, emphasising its efficiency for recycling and reuse. The magnetism of the metal nanoparticles decreases when they are incorporated with the biosorbents, while there is no significant influence on the separation and regeneration processes (Tung et al., 2018; Wang et al., 2014). The saturation magnetisation (Ms) of magnetic biosorbents is 16.00 emu/g, which is much lower than the pure iron oxide nanoparticles (Ms= 58.94 emu/g) (Wang et al., 2014). The magnetic properties of the magnetic biosorbents are positively correlated with the pyrolysis temperatures of the synthesis process. The magnetisation of biochar at 400 °C and 650 °C was 4.28 emu/g (not magnetically attracted) and 36.79 emu/g (magnetically attracted), respectively, which indicated that magnetic biochar should be pyrolysed at over 400 °C (Han et al., 2016). Therefore responsive magnetic biosorbents should be synthesised at higher temperatures. However, it also depends on the types of feedstock materials and types of impregnated metal /metal nanoparticles. Various types of modification methods, including pre-modification, post modification, microwave-assisted modification, and engineered modification, have been investigated for preparing magnetic biosorbents (Figure 2). The each types of modification method including advantage and disadvantages are critically evaluated in the following sub-sections.

2.1. Pre-treatment modification of biomass for synthesis of magnetic biosorbents

In recent years several scientific studies have conducted pre-treatment modification of biosorbents with magnetic nanoparticles (Akin et al., 2012; Alizadeh et al., 2018b; Cheng et al., 2012; Liu et al., 2010; Saravanan et al., 2012a; Zhou et al., 2018b). The materials prepared from pre-treatment (single step) modifications tend to show higher surface area, porosity, and superparamagnetic behaviour (Lian et al., 2014; Thines et al., 2017c; Yan et al., 2014b). Metal doped biosorbents induce metal-O/OH functional groups after pyrolysis in the pre-treatment method, which can influence the adsorption mechanism (Nisticò et al., 2018; Saravanan et al., 2012b). This is because the metal oxide nanoparticles being incorporated before the pyrolysis process exhibit oxidation and reduction potential through the utilisation of diverse species of metal nanoparticles in form of Fe^{2+} and Fe^{3+} (Akin et al., 2012; Alizadeh et al., 2018b; Cheng et al., 2012; Liu et al., 2010; Saravanan et al., 2012a; Tuna et al., 2013; Zhou et al., 2018b). Iron oxide reduction occurs during pyrolysis in the pre-treatment processes for iron-based biochar composites under a nitrogen atmosphere as described by Yang et al. (Yang et al., 2019a). As the pyrolysis temperature increased, amorphous carbon disappeared, while $FeCl_2$, Fe_3O_4, and Fe_3C appeared on the MB. The metal salt transfers to $Fe(OH)_3$ and FeO(OH) due to being hydrolysed with the biosorbents. During the pyrolysis process, some reducing components such as H_2, CO, and amorphous carbon could reduce FeO(OH) to Fe_3O_4 at high temperature. However, at high temperature pyrolysis (800 °C), few Fe_3O_4 could be transferred into Fe_3C due to reaction with carbon (Yang et al., 2016).

The transformation of the iron species during the hydrolysis and pyrolysis process could be explained by the following reactions,

$$FeO(OH) + H_2 \rightarrow Fe_3O_4 + H_2O$$

$$FeO(OH) + CO \rightarrow Fe_3O_4 + CO_2$$

$$FeO(OH) + C \rightarrow Fe_3O_4 + CO$$

$$FeCl_3 + H_2 \rightarrow FeCl_2 + HCl$$

Different metal or metal oxide nanoparticles were added into the raw biomass prior to further treatment (Figure 2). For example, Kataria et al. prepared magnetic nanoparticles incorporated into sawdust by the biogenic green synthesis approach (Kataria and Garg, 2018b). The sawdust was firstly mixed with iron nitrate ($Fe(NO_3)_3$) and then carbonised in a muffle furnace at 180 ºC for 12 h to introduce magnetic properties (Kataria and Garg, 2018b). Zhang et al. prepared magnetic biosorbents by immersing biomass into iron chloride ($FeCl_3$) solution (around 5 ppm concentration) before being pyrolysed at 600 ºC for 1h in the presence of N_2 (Zhang et al., 2013b). Zhu et al. (Zhu et al., 2014a) prepared magnetic porous carbon by immersing 20 g of hydro char materials into the prepared $FeCl_3$ solution (1.5 ppm) prior to drying and pyrolysis at 700 ºC for 1 h under N_2 flow.

2.2. Post-treatment modification of biochar for the synthesis of magnetic biosorbents.

Post-treatment (double step) modification is a conventional method and is less efficient than the single-step method in improving the surface area and sorption capacity due to cost and efficiency issues. Post-treatment modification involves preparing biosorbents and magnetic nanoparticles separately before being mixed. The mixture is shaken and cooled down after heating, prior to being washed and dried for storage and application. The most commonly used chemicals for magnetic modification are iron chloride, iron oxide, ferric nitrate, zero-valent iron, and metal-doped iron oxides (Figure 2). The mass ratio of magnetic nanoparticles and biochar generally ranges from 1:1 to 1: 40 (Han et al., 2015b; Harikishore Kumar Reddy and Lee, 2014; Song et al., 2014; Zhou et al., 2018b). For the preparation of biochar, the air-dried biomass is generally pyrolysed at 500-700 ºC for 1-2 h. Post-treatment modification requires more time and is expensive compared to the single-step modification method. However, it is possible to prepare different concentration ratios of magnetic nanoparticles and biosorbents, which is challenging to achieve in the pre-treatment procedure. For example, biomass waste was ground and positioned inside the furnace for heating at a high temperature (Hu et al., 2015). Then, the thermally modified biochar was mixed with ferric nitrate to allow the iron to be diffused in the biosorbents (Cope et al., 2014a). Feng et al. prepared magnetic biosorbents by Lotus steam-based raw biomass and iron chloride solution. After washing and drying, raw biomass was placed in a muffle furnace for pyrolysis at 500 ºC for 1.5 h. Then the prepared biochar was soaked in iron chlorine solution at a weight ratio of 1: 4 at 30 ºC for 2 h. The dried mixtures were then heated, washed, and dried for storage and application (Feng et al., 2018). Tuna et al. used ferrous and ferric chloride mixer solution with activated carbon (AC) at a mass ratio (AC: Fe) of 2: 1 for synthesis of magnetic biochar (Tuna et al., 2013). Dong et al. also used the double step modification method for preparing magnetic biosorbents using cornstalk as raw biomass (Dong et al., 2017b). The dried and ground cornstalk powder was placed in a tube furnace for pyrolysis at 500-700 ºC under limited oxygen conditions. The prepared materials were mixed with zero-valent iron and iron oxide solution (Dong et al., 2017b). Shi et al. also prepared magnetic biosorbents using zero-valent iron like Dong et al. (Dong et al., 2017a). Briefly, Phoenix tree leaves were used as a feedstock and pyrolysed at 700 ºC in a low oxygen environment. Then, the biochar was mixed with zero-valent iron (nZVI) at different mass ratios (1:1, 1:3, and 1: 5) to investigate their efficiency and usability. The higher Cr adsorption capacity was found at the 1:5

concentration ratio of zero-valent iron and biochar. Jiang et al. prepared magnetic biochar using palm biomass and this was pyrolysed at 600 ºC for 8 h. Then the powder biochar was dispersed in 30 ml 1M $FeCl_3$ solution for 3 h (Jiang et al., 2018b). Corncob husk and sugarcane bagasse were also investigated for pyrolysis and preparation of magnetic adsorbents. They were pyrolysed in a muffle furnace prior to being immersed in a $FeCl_3$ solution to generate Fe-coated biochars (Farooq et al.; Montero et al., 2018). Similarly, the peanut and rice straw were pyrolsed prior to being immersed in the $FeCl_3$ solution with magnetic stirring (Pan et al., 2015). Both pre and post-treatment modification methods are used in Baig et al.'s study on the preparation of magnetic biosorbents: (1) Kans grass was first pyrolysed into biochar and fabricated with Fe_3O_4, and (2) Fe_3O_4 was loaded onto the dry biomass by chemical co-precipitation prior to being pyrolysed (Baig et al., 2014b).

2.3. Synthesis of mineral-supported biosorbents

The typical methodology to prepare mineral-supported magnetic biosorbents is illustrated in Figure 2. Various low cost and naturally occurring minerals are used to synthesise magnetic biosorbents. Wang et al. (Wang et al., 2015f) prepared an adsorbent by mixing well-dispersed hematite solution (sonicated for 30 min) with pinewood as a feedstock material for 2 h and then oven-dried. The hematite-mixed biomass was pyrolysed under N_2 at 600 °C (Wang et al., 2015f). Wang et al. (2015d) incorporated Loblolly pine (Pinus taeda) wood with $MnCl_2 \cdot 4H_2O$ solution. The mixture was then oven-dried prior to being pyrolysed in a tube furnace at 600 ºC for 1 h under a nitrogen environment (Wang et al., 2015d). The authors also prepared magnetic biosorbents by mixing pine biochar and synthesised Birnessite solution, which was then agitated for 2 h. The suspension was boiled for 20 min, followed by the addition of concentrated HCl to the solution. The suspension was dried overnight at 80 ºC. They found that the mineral-supported biochar had better adsorption capacity than metal-doped biochar for As and Pb. However, the synthesis process is more complicated compared to other methods (Wang et al., 2015d).

2.4. Microwave-assisted magnetic biosorbents synthesis

Another emerging and efficient method for preparing magnetic biosorbents or biochar is with the assistance of microwave technology. The preparation of magnetic biosorbents through the microwave requires only 5-10 minutes, which is much lower than the conventional muffle furnace assisted method. Additionally, this process is easy to operate compared to other existing synthesis methods. However, it is difficult in terms of specific customisation for tailoring a synthesis method based on particular requirements. This idea was originally propounded around 42 years ago by Ramasahayam et al. (Ramasahayam et al., 2015). The microwave-assisted magnetic biochar demonstrated 1-10 times superior adsorption capacity, higher surface area, and pore volume than conventionally prepared biosorbents (Thines et al., 2017b; Wang et al., 2013a). It is an example of a single step synthesis method (Figure 2). The microwave and conventionally prepared magnetic biosorbents exhibited excellent ferromagnetic properties with a magnetisation of 8.16 and 4.20 emu/g, respectively (Mubarak et al., 2016). Thus, the microwave-assisted modification could be an effective alternative to conventional pyrolysis. Various raw materials like bamboo, fruit peel, and chitosan can be modified with magnetic nanoparticles in a microwave for the synthesis of magnetic biosorbents. The modification required 600-800 W power (2.45 GHz) for 5 to 30 minutes under a nitrogen environment. Wang et al. (Wang et al., 2013a) synthesised Co-Fe magnetic biochar through the assistance of microwave technology. The prepared biochar was introduced into $FeCl_3$ and $Co(NO_3)_2$ mixture solution at room temperature and then heated in a microwave (Wang et al., 2013a). In another study (Mubarak et al., 2016), the dried biomass was crushed and sieved before being impregnated with ferric chloride. The pyrolysis of the biomass was carried out in a microwave muffle system (Mubarak et al., 2016). Ni-doped bamboo charcoal was prepared using a similar procedure (Thines et al., 2017b). The $NiCl_2$ solution was mixed with the original biochar prior to ultra-sonication at room temperature. Then, the biochar was separated from the mixtures and placed in a modified microwave heating under a nitrogen flow (Thines et al., 2017b).

2.5. Hydrothermal method for synthesis of magnetic biosorbents

Hydrothermal carbonisation is a thermochemical process, where the feedstock materials are mixed with water at a temperature of 100−350 °C. The use of water helps to hold gases in the water medium and reducing CO_2 emission but low surface area and porosity result from the hydrothermal carbonisation. Biochar production by the hydrothermal process is less common than conventional carbonisation processes. About 90% of sulfur, silicon, calcium, iron, magnesium, phosphorus, potassium and other metals from feedstock materials can be removed by hydrothermal carbonisation. Thus, the hydrothermal process is not receiving much attention for the preparation of adsorbents from feedstock materials (Gao et al., 2016; Kambo and Dutta, 2015; Varma, 2019). Few studies have investigated the hydrothermal method. Lian et al. (Lian et al., 2014) synthesised iron-loaded biochar using a hydrothermal reaction of iron chloride and microalgae as a feedstock material. The microalgae were collected, dried and ground to a powder prior to being mixed with different amounts of iron for reaction in an electric oven. The mixture was centrifuged to separate the iron incorporated biochar. The obtained Fe coated biochar was washed with distilled water and ethanol solution to remove all debris matter. Finally, it was dried in an oven (Lian et al., 2014). The authors also used the hydrothermal magnetisation of the carbonised corncob to synthesize the derived magnetic biosorbents. The carbonised materials and $FeCl_3 \cdot 6H_2O$ were dispersed in deionised water in the presence of NaOH. After cooling, the magnetic biosorbents were collected by a magnet, washed with water until neutral and dried before storage. In hydrothermal magnetisation, the yield of the final magnetic activated carbon obtained was 75%, which was calculated based on the initial amount of carbon present (Lian et al., 2014). Hydrothermally synthesised magnetic biosorbents can be used to remediate organic and inorganic contaminants (e.g., microcystin-LR, methylene blue) (El Mouzdahir et al., 2007; Lian et al., 2014; Sivashankar et al., 2014a) while the synthesis process is a bit complex. Only a few studies have been conducted using this method.

2.6. Synthesis of engineered or tailored magnetic biosorbents

Engineered magnetic biosorbents constitute an innovative approach to tackling wastewater treatment, and are prepared directly from plant tissues rich in certain types of metals through bioaccumulation (Salt et al., 1995). The target element can be accumulated in biomass, which could be further used to produce engineered biochar. Iron or other nanoparticles may accumulate in plants and be directly used to produce magnetic biosorbents without any further treatment or modification. Magnetic biosorbents can be developed through bio-accumulation of metals or desired nanoparticles in plant tissue, which can be easily used without complex modification before or after pyrolysis. It appears to be a promising technology but currently is still in the experimental phase. In this process, the plant uptakes the supplied metal ions or other chemicals, which helps to develop multifunctional chemical properties through pyrolysis (Tan et al., 2016a). There is a limited number of reports on these processes for removing contamination from water. Ying et al. reported increasing bioaccumulation of manganese in tomato plants (8% Mn of total dry biomass of tomato plant) in a greenhouse, which improved the physical and chemical properties and adsorption capacity of the final product. The adsorption capacity increased around 5-6 times compared with standard activated carbon for phosphorous remediation (Yao et al., 2013b). Tang et al.

reported that iron bioaccumulated plant tissues had also improved physicochemical characteristics and increased adsorption capacity of Chlorpyrifos. Iron nanoparticles are bioaccumulated (1% w/w) by allium fistulosum (Welsh onion) which enhanced the adsorption capacity of organophosphorus insecticide (Eevers et al., 2017; Tang et al., 2017).

2.7. Comparison of different synthesis methods for magnetic biosorbents.

The pre-pyrolysis modification mixes biomass feedstock with metal nanoparticles, prior to thermal treatment in the presence of nitrogen for a stipulated time period. The post-pyrolysis modification generates biochar first from biomass, followed by impregnation with metal nanoparticles (Figure 2). Conventional furnace heating and microwave-assisted pyrolysis can be used for the preparation of magnetic biosorbents (Baig et al., 2014b). The conventional method is most widely used for nutshell modification, while high quality synthesised magnetic biosorbents are prepared through microwave (Thines et al., 2017c). The conventional method is more efficient and flexible for tailored functionality for particular contamination removal, as it is possible to manipulate modification factors, including gas flow and temperature. The single-step modification method is the commonly used conventional strategy due to the production of versatile functionality on the adsorbents' surfaces. Oxidation-reduction of metal nanoparticles or their derivatives can occur during pyrolysis in the single-step method, which helps the development of more diverse functional groups than the post-treatment method. This is due to the oxidation-reduction of pretreated magnetic nanoparticles. The magnetic biosorbents prepared from single and double step methods show significantly different properties due to the presence or absence of oxidation-reduction of metal nanoparticles. The microwave-assisted biosorbents and mineral-supported magnetic biosorbents can also be considered as a one-step modification method. Mineral supported biosorbents have a comparatively low production cost compared with conventional magnetic nanomaterials modified biosorbents. Hydrothermal carbonisation is a thermochemical heating process under water conditions at a heating temperature of 100−350 °C. Hydrothermal carbonisation produces low greenhouse gas emissions but it generates low surface area and porosity, which limits its application. Biochar production by the hydrothermal process is comparatively less common than conventional carbonisation processes. It is evident that engineered magnetic biochar appears to be an effective, green, and sustainable synthesis method for preparing the desired adsorbent that should be assessed by future research activities.

3. Application of magnetic biosorbents for the removal of organic and inorganic contaminants

Magnetic biosorbents is an effective adsorbents for inorganic and organic contaminants removal (Han et al., 2015b; Rajapaksha et al., 2016; Yoon et al., 2017). The removal of contaminants from water via the adsorption process depends on the physical and chemical properties of adsorbents including surface area, porosity, and sorption parameter including pH, initial concentration, adsorbent dose, and contact time (Mehta et al., 2015; Zhang et al., 2013b). The synthesis method, characteristics of MB, background electrolytes, sorption conditions, isotherms, and kinetics for contaminants removal are highlighted in Table 1. The effects of the physicochemical properties, adsorption condition, and solution chemistry of magnetic biosorbents on the adsorption of the contaminants were discussed in the following section to understand the sorption mechanisms and behaviours.

3.1. Sorption of inorganic cations by magnetic biosorbents.

Magnetic biosorbents are reported to remediate wide ranges of cations including Pb^{2+}, Cd^{2+}, Cr^{6+} and Cu^{2+} from wastewater (Table 1). For example, Han et al. (Han et al., 2015b) prepared magnetic activated carbon, and magnetic biosorbents for Pb removal, where the adsorption capacities ranging between 11.6 mg/g to 35.7 mg/g. Novel multifunctional magnetic biosorbents can remove more than 97% cobalt from real wastewater via the formation of bis and tris complexation with a carboxylic acid functional group (Gao et al., 2017). Removal efficiencies reached 99.4%, 66.7%, and 99.1%, for Pb^{2+}, Cr^{6+} and Cu^{2+} respectively on zero-valent iron modified biosorbents via complexation with −OH/–COOH functional groups or delocalised π electrons. Iron-based functional group can also adsorb heavy metals via transferring into $Fe–O–M^{n+}$ complexation (Zhu et al., 2017).

The removal efficiency by magnetic biosorbents are generally higher than its pristine materials. For example, Yan et al. synthesised magnetic biosorbents with ZnS nanocrystals which exhibited about 10 times greater adsorption capacity than the unmodified biosorbents (Yan et al., 2015b). Iron chloride modified magnetic biosorbents improved adsorption capacity for mercury (Hg) via a coordination bond with the C=O group (acting as an electron acceptor) and lattice oxygen (Yang et al., 2016). Zinc borate pre-treated biomass enhanced the biochar yield and promoted an abundance of the surface oxygen group that enhanced nickel adsorption capacity up to 3-10 times compared with the unmodified biochar (Liu et al., 2014). Similarly, manganese modified wood-derived biosorbents improved adsorption capacity 2.1 times for lead, 2.8 times for copper, and 5.9 times for cadmium compared with the pristine biochar due to more oxygen-containing functional groups being induced and a larger surface area (Wang et al., 2015a). In other studies, the adsorption capacity of lead by manganese induced biosorbents improved up to 2 to 20 times due to more oxygen-containing functional groups (Wang et al., 2015b; Wang et al., 2015d). Additionally, nitrogen-induced magnetic biosorbents also improved adsorption capacity for lead (893 mg/g) within a short equilibrium time (less than 10 minutes) (Ling et al., 2017) via surface coordination of

Pb^{2+} and C═O or O═C–O, and the presence of aliphatic N. Amorphous MnO_2 modified biosorbents demonstrated an adsorption capacity of 248.0 mg/g of lead and 45.8 mg/g of Cd via Pb–O / Cd-O or hydroxyl binding and ion exchange reactions, which were the primary separation mechanisms (Liang et al., 2017). This suggested that engineering functional groups of biosorbents can achieve higher adsorption capacity for the removal of cations from wastewater (Ling et al., 2017).

The removal efficiency of Cd by magnetic biosorbents was compared with various sorbents (Figure 3), which showed that magnetic biosorbents are superior to most of the other conventional adsorbents. The mean value of sorption capacity for magnetic biosorbents is comparatively higher than activated carbon, minerals, polymers, plant wastes, nanomaterials and nanocomposites. Microbial waste also shows good adsorption capacity due to their complex structures and diverse elemental compositions which helps to produce more active sites to adsorb ionic contaminants. The high surface area and easy separation of magnetic biosorbents make them effective for the elimination of Cd from the wastewater. The relevant information reported in the literature on using magnetic nanoparticle modified biosorbents for Cd removal is summarised in Table SM2. A moderate positive correlation was found between wt. % of hydrogen and adsorption capacity for Cd removal, whereas a limited negative correlation has been observed between cadmium adsorption and wt. % of metal nanoparticles due to competition with cadmium ions in the adsorption medium. Accordingly, the impregnation of metal nanoparticles should be controlled in the magnetic biosorbents to avoid competition on the adsorbent's active sites. The relationship between physicochemical properties of magnetic biosorbents and sorption capacity of cadmium were analysed using SPSS software as shown in Table SM4.

The cations can be sorbed through chemisorption, surface complexation, and co-precipitation (Wang et al., 2018a). Additionally, cations could be attracted and bonded through ion exchange, hydrogen bonding, chemisorption, chelation, or complexation (Sud et al., 2008a). The metal induced multifunctional groups in magnetic adsorbents can influence the sorption mechanisms of cations. The presence of iron and other metals and salts in industrial waste and magnetic biochar contributed more diverse functional groups apart from that from raw biomass derived biochars. The primary functional groups that contribute to cations removal include carboxyl (-CO), ester (-COO), metal (Fe/Ni/Ti/Zr - O/OH), amine ($-NH_2$) groups (Chand et al., 2014; Rajapaksha et al., 2016; Yang et al., 2019a; Yang et al., 2019b; Yoon et al., 2017; Yoon et al., 2019). Hydroxyl group, carboxyl group, amino group, carboxylic acid and M-O/OH are mainly responsible for an ion exchange reaction whereas carboxylic acid and carboxyl group are responsible for electrostatic interaction (Table 2). Carboxyl group, hydroxyl group, amino group, carboxylic acid, and amino group can contribute to adsorption of cations through complexation on the surface of the adsorbents (Table 2). Carboxyl, hydroxyl, and carbonyl are responsible for the chelation process (Yang et al., 2019d). It has been reported that the presence of γ-Fe_2O_3 nanoparticles increased adsorption capacity of catalyst doped (e.g. $ZnFe_2O_4$, $CuFe_2O_4$) magnetic carbon composites than just magnetic biochar (Wang et al., 2018a). Similarly, the presence of multifunctional groups due to using γ-Fe_2O_3 instead of raw iron oxide nanoparticles improved adsorption capacity from 31.80 to 167.22 mg/g for Cd (Wang et al., 2018a). Generally, nanoparticles incorporating biochar have an inferior adsorption capacity (less than 100 mg/g) compared with magnetic biosorbents which are further modified at multiple stages for cations removal (Kataria and Garg, 2018a; Liu et al., 2009; Zhang et al., 2017c). The utilisation of Fe_3O_4@layer double hydro oxide@bionanocomposite (magnetic biocomposite) improved the adsorption capacity to 258.00 mg/g (Dinari and Tabatabaeian, 2018) whereas the adsorption capacity for magnetic oak wood biochar was 66.22 mg/g due to variation of functional group (Mohan et al., 2014b). Similarly, magnetic EDTA/Chitosan/TiO_2 had an adsorption capacity of 209.21 mg/g, whereas it was only 51.00 mg/g when using Fe_3O_4 coated sawdust carbon (Kataria and Garg, 2018a). The incorporation of TiO_2, EDTA, or other chemicals with magnetic biochar introduced some additional functional groups that favored adsorption. Likewise, ferromanganese binary magnetic biochar had an adsorption capacity of 101 mg/g (Zhou et al., 2018a) while $MnFe_2O_4$ biochar had an adsorption capacity of 181.49 mg/g due to Mn-doped iron oxide that induced the Mn-O functional group (Wang et al., 2018b). nZVI modified biosorbents from cornstalks (magnetic biosorbents) has reported as effective adsorbents for single and mixed metals solution and equilibrium adsorption capacities reach about 195, 162 and 110 $mg \cdot g^{-1}$ for Pb, Cu and Zn divalent ions at neutral medium (Yang et al., 2019a). The possible reactions mechanism of the divalent ions on the magnetic adsorbents were adsorption, reduction and precipitation (Yang et al., 2018; Yang et al., 2019a). The role of iron nanoparticles to adsorption and reduction of heavy metals highlighted from Yang et al.(Yang et al., 2018) .

$M^{2+} + Fe0 \rightarrow M - Fe^{2+}$ (adsorption)

$M^{2+} + Fe0 \rightarrow M + Fe^{2+}$ (reduction)

(Where, $M^{2+} = Pb^{2+}$, Cu^{2+}, or Zn^{2+})

The sorption of cations by magnetic biosorbents can be influenced by solution pH. Solution pH influences adsorption performance of cations through protonation/deprotonation of surface functional groups which demonstrated that electrostatic interaction is one of the prominent mechanisms for cations adsorption. The interrelationship of pH, point of zero charge, background electrolytes, and adsorption capacity of cadmium was profoundly studied by Naidu et al. (Naidu et al., 1994). However, point of zero charge of magnetic biosorbents may decrease due to the magnetisation of biosorbents (Han et al., 2015b). At $pH < pH_{ZPC,}$ the surface of magnetic biosorbents are protonated which makes the surface electropositive and leads to the electrostatic attraction for anions but electrostatic repulsion for cations. At pH >pH_{ZPC}, the adsorbents are covered by the hydroxyl group on the magnetic biosorbents surface that leads to the electrostatic attraction for cations but electrostatic repulsion for anions. The presence of co-existing cations and anions can be used for controlling the electrostatic attraction and repulsion, and competing effect of similarly charged ions. The electrostatic attraction or repulsion can be controlled by forming a monolayer of co-existing cations and anions on the magnetic biosorbents. However, the effect of pH and pH_{ZPC} for removal of cations and anions is highly interconnected whereas the effect of solution pH is insignificant for adsorbing non-ionic organic contaminants. The electrolyte's effect is slightly interrelated (Lalhmunsiama et al., 2017) where the frequently used background electrolytes are 0.01 M $NaNO_3$, $MgCl_2$, 0.01 M Na, and 0.01 N Na_3PO_4, NaCl, Na_2SO_4, $NaNO_3$ for improving adsorption capacity and ionic strength of the solution.

3.2. Sorption of inorganic anions by magnetic biosorbents

The removal of major anions from water by magnetic biosorbents is summarised in this section which indicated higher adsorption capacity of magnetic biosorbents than its raw biomass or pristine biochars (Table 1). For example, the adsorption capacity of phosphate by magnetic biochar is much higher (290 – 887 mg/g) than the pristine biochars due to the enrich functional groups and nano-flakes on the biosorbent's matrix after modification with Al/Mg/Fe metals (Fang et al., 2014; Fang et al., 2015; Jung et al., 2015; Yao et al., 2013a; Yao et al., 2013b). The adsorption capacity of nitrates is also high by magnetic biosorbents, which was reported as 95 mg/g on $MgCl_2$ treated biosorbents (Zhang et al., 2012). Similarly, biosorbents modified with zinc nitrate/ iron/ calcium agents also improved Cr(Yang et al.) and Cr(VI) sorption capacity to almost double that of the pristine biochars (Agrafioti et al., 2014; Gan et al., 2015). The adsorption capacity for Cr(VI) and As(V) are higher on the magnetic biochar than the non-magnetic biochar (Agrafioti et al., 2014). Wen et al. synthesised magnetic porous carbonaceous materials which showed higher adsorption capacities for As(V) (38.03 mg g^{-1}) and Cr(VI) (21.23 mg g^{-1}). Zhong et al. synthesised magnetic biochar composite via single-step microwave pyrolysis of iron sulphate and rice husk (feedstock materials) to remediate Cr(VI) from water. The magnetic biochar exhibits 3.2 and 11.7 times higher sorption and reduction capacity of Cr(VI), due to enhanced surface area, porous graphitic structure, pore volume and reactive magnetite to sorption or reduction of Cr(VI) (Zhong et al., 2018). Moreover, the positively charged magnetic adsorbents can be used to separate anionic dye and humic acid (Wen et al., 2017).

Sorption of As ($H_2AsO_4^-$ and $HAsO_4^{2-}$) by magnetic adsorbents is used as a case example for understanding the relationship between different factors and sorption capacity. A comprehensive list of adsorption capacity for arsenic by magnetic biosorbents and the physiochemical properties are highlighted in supplementary materials as Table SM3 to represent the overall mechanisms and tailored development of magnetic biosorbents. Magnetic biosorbents showed high adsorption capacity for As ranging from 5 to 500 mg/g (Liu et al., 2010; Zhang et al., 2013b) with different types of metal oxide incorporated into biosorbents. A zero-valent iron (nZVI) modified biochar has an adsorption capacity of 124.50 mg/g which is higher than the other metal oxide impregnated biosorbents (Baig et al., 2014a; Wang et al., 2017). However, nZVI can be easily oxidised and form aggregation after physical contact with several (mostly organic) contaminants whereas there are no such concerns for metal oxide nanoparticles. Thus, a metal oxide is more applicable than zero-valent iron when incorporated into biosorbents (Lunge et al., 2014). In another study, sawdust modified with $MnFe_2O_4$ exhibited an adsorption capacity rising to 507 mg/g for As (Podder and Majumder, 2015). The metal doping iron oxide is more efficient than just raw metal oxide nanoparticles for preparing magnetic biosorbents (Podder and Majumder, 2015; Zhang et al., 2017c). The correlation of magnetic biosorbent properties with As sorption capacities was analysed using SPSS software, which is shown in Table SM4. Positive correlation was found between wt % of Fe, Wt. % of H, pore size and sorption capacity of arsenic. The correlation values indicates the wt. % of iron content positively influencing arsenic adsorption without competition with arsenic on the active sites of the adsorbents, due to opposite charges of metal nanoparticles and arsenic.

A wide variety of materials are compared for removing arsenic from wastewater, including biochar, nanocomposite, biosorbents, metal oxide, minerals, laterite soil, residue materials, red mud, and other low-cost adsorbents (Terán Hilares et al.; Wang and Tsang, 2013; Yadanaparthi et al., 2009a). Activated carbon, polymer, industrial waste materials and thermally and chemically (acid and base) modified biochar were widely used but exhibited lower removal efficiency. Conversely, metal oxide nanoparticles, nanocomposites, modified iron oxide nanoparticles, and magnetic biochar demonstrated higher removal efficiency for arsenic removal compared with other sorbents (Figure 4). Metal nanocomposites and nanomaterial derived adsorbents showed the highest adsorption capacity for arsenic removal, as shown in Figure 4. The sorption capacity of carbon nanotubes modified with different metal oxides was below 1 mg/g (Addo Ntim and Mitra, 2011; Ntim and Mitra, 2012; Tawabini et al., 2011). Similarly, minerals like gibbsite, kaolinite, goethite, hematite, and zeolite can sorb As with their adsorption capacity below 7 mg/g (Altundoğan et al., 2000; Li et al., 2007; Singh et al., 1988). Moreover, for most studied biochars, the sorption capacity was near 15 mg/g or below 20 mg/g for As (Cope et al., 2014b; Mohan et al., 2007; Samsuri et al., 2013; Wang et al., 2015d; Zhang et al., 2013b). In contrast, polymer functionalised nanocomposite could adsorb between 10 to 25 times more than biomass, biochar, waste materials and carbon-based nanomaterials (CNTs) (Nie et al., 2015). For metal oxide nanoparticles, the adsorption capacity was found to be around 5-20 mg/g whereas modified iron oxide showed much higher adsorption capacity ranging from 50 to 250 mg/g for arsenic removal (Daus et al., 2004; Lenoble et al., 2005; Peng et al., 2017; Xu et al., 2012). This could be due to the development of more functional groups at the time of modification with other nanomaterials, photo-catalysts, graphene, acids, and bases (Chowdhury and Yanful, 2010; Kumar et al., 2014; Lin et al., 2012; Lunge et al., 2014; Luo et al., 2012; Yang and Yin, 2017; Yu et al., 2015). Generally, magnetic biosorbents show higher adsorption capacity for arsenic removal (Baig et al., 2014a; He et al., 2018; Zhang et al., 2017c).

The complexation and electrostatic interaction mechanisms are involved in anions removal. For example, complexation of phosphate and magnetic biochar as $[(Fe–O)_2\text{-}PO_2H]^{–}$, $[Fe–O–PO_3H]^{–}$ and $[Fe–O–PO_3]^{2–}$ at acidic, neutral and alkali condition were reported that can adsorb via (Karunanayake et al., 2019). At the same time, precipitation and co-ordination bond can immobilise arsenic from the water solution. The carboxyl group, amino group, carboxylic acid, and thiol amino group are the main functional groups for adsorbing the arsenic species from water (Table 2). Electrostatic interactions are prominent for the removal of anionic contaminants at low pH, whereas high pH is suitable for removal of cationic contaminants (cadmium, lead, copper, and zinc) via electrostatic interaction (Nisticò et al., 2018; Sun et al., 2015; Zhou et al., 2014). For example, adsorption capacity was low at high pH, whereas it was high at low pH for anions (Wang et al., 2015f; Zhang et al., 2013b). Zhuo et al. (Zhou et al., 2017) observed the effect of background electrolyte at the point of zero charges where the effect is not adequately explained. However, it has been found that the adsorption of anions is lower in the presence of co-existing anions than the cations (Zhou et al., 2017). These phenomena occur due to electrostatic repulsion between the adsorbents and the anions. Thus, the phenomena can only be explained at higher or lower pH than the point of zero charges (pH_{pzc}). When $pH < pH_{pzc}$, the surface of the biosorbents is positively charged, which favours arsenic adsorption. Presence of anions may decrease the adsorption capacity due to the competition with anions whereas the presence of cations may increase the adsorption capacity due to monolayer formation of cations on the surface of the negatively charged adsorbents. Similarly, When $pH > pH_{pzc}$, the presence of cations increased the adsorption capacity of arsenic by forming a monolayer of positively charge cations on the adsorbents that can bind anions through the electrostatic interactions whereas the presence of anions may have a decreasing anions adsorption.

3.3. Sorption of organic contaminants by magnetic biosorbents

Several organic contaminants present in wastewater, including dyes, pesticides, and emerging organic contaminants can be adsorbed by magnetic biosorbents (Rahman et al., 2014). The magnetic biosorbents have the potential to efficiently remediate organic contaminants by adsorption and catalytic degradation (Chen et al., 2011; Sun et al., 2015; Zhang and Gao, 2013) while only a few studies have investigated the removal of organic contaminants by magnetic biosorbents (Table 3). The electrostatic interactions, ion exchange, π-π interaction, interparticle diffusion hydrophobic interactions, van der waals interactions, and hydrogen bonding are the major adsorption mechanisms for removal of organic contaminants including PFOS (perfluorooctanesulfonic acid), tetracycline, doxorubicin, levofloxacin, ciprofloxacin (CIP), sulfamethoxazole (SMX), and doxycycline (Du et al., 2014; Zeng et al., 2018a; Zeng et al., 2018b) (Li et al., 2017).

Magnetic biosorbents can remediate organic contaminants via either sorption or catalytic activity, including acid orange 7, methylene blue, nitrophenol, methyl orange, pentachlorophenol, tar, trichloroethylene, other pesticides, antibiotics and emerging contaminants (Devi and Saroha, 2014; Devi and Saroha, 2015; Han et al., 2015a; Kumar et al., 2017; Li et al., 2019b; Pi et al., 2015; Quan et al., 2014; Tan et al., 2016a). For example, Oh et al. synthesised zero-valent iron (nZVI) induced biochar for 2,4-Dinitrotoluene and 2,4-dichlorophenol separation via hydrophobic interaction that can be further reduced by the magnetic biosorbents (Oh et al., 2016). Similarly, Yan et al. synthesised nZVI induced biochars utilised for trichloroethylene degradation via activation of persulfate to generate SO_4^-, with its oxygen-containing functional groups including carboxyl (—COOH), hydroxyl (—OH) and Fe^{2+}/Fe^{3+} within 5 min (Yan et al., 2015a). Calcium salt pre-treated biosorbents also enhanced adsorption capacity for acid blue 74 and reactive blue 4 due to increased micropore and total pore volume (Aguayo-Villarreal et al., 2014). Similarly, zinc chloride ($ZnCl_2$) modified sewage sludge also enhanced the adsorption capacity of benzene derivatives due to improved micropore, mesopore, and specific surface area (Kong et al., 2014). Fe_3O_4 modified biosorbents synthesised via a co-precipitated method can also adsorb crystal violet nearly 20 times higher than the pristine biosorbents. A microwave-assisted magnetic biochar exhibited higher adsorption capacity (265 mg/g) of methylene blue due to its high specific surface area and hydrophobic interaction (Mubarak et al., 2014a). Magnetic activated carbon also exhibited high adsorption capacity (14 mg/g) for Sunset Yellow (SY) (organic dyes) (Cazetta et al., 2016). Thines et al. used magnetic biosorbents derived from sawdust for sulfamethox azzole remediation, with the removal capacity being 8.73 mg/g (Thines et al., 2017c). Sun et al. observed that magnetic biochar could adsorb up to 349.40 mg/g crystal violet (Sun et al., 2015). Mubarak et al. claimed that synthesis of palm oil-derived magnetic biosorbents with microwave assistance could remove up to 99.9%

methylene blue (Mubarak et al., 2014b). Solid waste modified with ferric chloride at a mixing ratio of 2:1, demonstrated an adsorption capacity of 259.25 mg/g for methylene blue at pH 7 and contact time of 4.5 h. This sorption procedure followed Pseudo-second-order kinetics and the Langmuir isotherm model (Theydan and Ahmed, 2012). Similarly, co-precipitated magnetic biosorbents can co-separate phosphates and methylene blue simultaneously at pH 7.0 with a contact time of 4.5 h but no significant competition between the sorbates (Chen et al., 2011). Mixed metal-biochar composites via pyrolysis at 700 °C by using red mud (RM) and lignin, to induce catalytic degradation and reductive properties of metal and hydrophobic properties of porous lignin to remediate heavy metal(loid)s, methylene blue, para-nitrophenol and pCBA. The silicon, iron, titanium induced functional groups from redmud and hierarchical porous carbon structure contributed to the adsorption, reduction and catalytic reaction of the selected organic and inorganic contaminants (Cho et al., 2019).

Metal nanoparticles modified biosorbents have high catalytic, oxidative/reductive and sorption properties due to the presence of metal nanoparticles, oxygen-containing functional groups (O-H, COOH-), and induced new functional groups as metal–O/OH. As a result of the multifunctional properties of magnetic biosorbents, they have become one of the advanced materials for remediation of organic contaminants from wastewater. Carboxylic acid and carboxyl group are mainly responsible for ion exchange and electrostatic interaction, whereas the C-X [where X-Halogen] functional group is responsible for the hydrogen bonding. However, hydrophobic interaction is the prominent sorption mechanism for removal of organic contaminants. Thus, biomass is pyrolysed at higher temperatures to develop the hydrophobic characteristics of the biochar that can efficiently bind organic contaminants from water. Hydrophobicity of the biosorbents depends on the aromatic structure of biosorbents that are suitable for removing organic contaminants. Han et al. (Han et al., 2015b) claimed that the adsorption capacity of organic contaminants correlated mainly with surface area, while inorganic cations and anions did not follow such a strong correlation with the surface area. Some organic contaminants (e.g., PFOS) have charges that could be adsorbed partially with electrostatic interaction and ion exchange at low or high pH. Additionally, solution pH and electrolyte concentration (except above 1 M) insignificantly affect the removal of organic contaminants. To be specific, for electrolytes below 1 M, the adsorption capacity generally does not alter significantly, but above this concentration, the adsorption capacity could be reduced (Afzal et al., 2018).

However, In real wastewater treatment, effluents may contain various heavy metal(loid)s, organic contaminants, dissolved organic carbon (DOC), salt ions including chlorine, sulphate and nitrate which may compete to adsorb active sites on the adsorbents (Huang et al., 2014; Lei et al., 2017; Liao et al., 2018). Similar charges ions are generally compete with similar form of ions, where electrostatic and ion exchange reaction is principle sorption mechanism. For example, magnetic biochars exhibited a high sorption performance for Cd^{2+}, Cu^{2+}, and Zn^{2+} removal despite having high selectivity for Cu^{2+} due to its high affinity with COOH- and -OH (Son et al., 2018). Similarly, The effect of coexisting anions competing with phosphate adsorption sites was: $F^- > NO_3^- > Cl^- > SO_4^{2-}$ on the zirconium and iron modified activated carbon (Xiong et al., 2017). However, very few articles discuss about the selectivity of magnetic biosorbents in the real wastewater treatment or environmental condition, which should be further study in details to come up with complete conclusion.

4. Possible management of magnetic biosorbents after application for wastewater treatment

Biosorbents, iron oxide and its derivatives and the removed contaminants themselves can be sources of environmental contamination if they are not managed and disposed of appropriately. The details for the safe management of magnetic biosorbents are discussed in more detail in the following sections.

4.1. Recycling of magnetic biosorbents

The capacity for recycling magnetic biosorbents is still under investigation. Only a few studies have reused magnetic biosorbents 3-7 times without any significant reduction of the adsorption capacity at lab-scale (Wang et al., 2015c). Different acids and bases (NaOH, HCl, H_2SO_4, EDTA, HNO_3, $Ca(NO_3)_2$, $NaNO_3$) are used as reagents for desorbing contaminants from magnetic biosorbents. For example, arsenic can be desorbed from magnetic biosorbents up to 70-90% with 0.5 M NaOH (Baig et al., 2014b). Similarly, cation-loaded magnetic biosorbents (Fe_3O_4@orange peel) can be recycled using 0.1 M HNO_3. Low concentrations of acid or base (0.1-0.2 M) are usually used for desorption of contaminants from magnetic biosorbents. The recovery of magnetic biosorbents using inorganic solvents is summarised in Table 4. Magnetic biosorbents are also regenerated by organic solvent. Wang et al. used acetic acid, water, and EDTA as well as 0.1 M HCl in order to understand the efficacy of recycling magnetic biosorbents. Higher desorption efficiency and less iron leaching were found when using EDTA (Wang et al., 2015c). Reduced leaching of iron content from the magnetic biosorbents retains the magnetism of the MB, which could be further used for separation using magnetism (Wang et al., 2015c). The efficacy of recycling magnetic biosorbents using different organic solvents was examined by Reguval et al. (Reguyal et al., 2017b), which revealed that methanol, ethanol, and acetone have higher regeneration capacity by separation of organic contaminants from the biosorbents.

However, the application of magnetic biosorbents for real wastewater treatment is limited. One pilot-scale application of magnetic clay based adsorbents for real wastewater treatment was reported by Salinas et al. (Salinas et al., 2018). The authors proposed an operating diagram for magnetic separation, which includes a rotating magnetic dump, wastewater inlet, outlet, and magnetic adsorbents separator (Figure SM2). In general, the fixed-bed column is used due to the complex sedimentation process. However, for better contact between contaminated water and adsorbents, the adsorbents should be freely floating into the wastewater. It is quite challenging for sedimentation of floating adsorbents after the adsorption processes. The sedimentation is a kind of simple solid-liquid separation process where the solid is to be deposited on the bottom of the tank and afterward collected from the bottom of the tank (Edzwald, 2010). Sedimentation is not an efficient technique due to the longer time required, which also depends on particle size, density of adsorbents, and distance between adsorbents and bottom of the tank. Magnetically modified clay can be easily separated with the assistance of a high magnetic field that can reduce sedimentation time by up to 90% (Salinas et al., 2018). The major drawback of regeneration of magnetic biosorbents is the introduced organic and inorganic solvents used to separate contaminants from the surface of the adsorbent that may lead to secondary waste. Mixture of solvents and contaminants can be a serious environmental issue until its safe disposal and proper management.

4.2 Fate of magnetic biosorbents

The metal-based nanoparticles can go through various transformation behaviors once introduced into the water and terrestrial environments (Lombi et al., 2012). For example, iron based nanoparticles including zero valent iron nanoparticles emerged as environmental concern due to physical, chemical and biological transformation, which is influenced by natural organic matter, pH, coexisting cations and anions, dissolved oxygen, biotic and abiotic system in real wastewater system. Biotic and abiotic system including bacteria, fungi, algae, fish may be affected by the toxicity of iron based nanoparticles due to formation of reactive oxygen species from iron nanoparticles. However, surface coating may reduce toxicity of metal nanoparticles by reducing transformation of nanoparticles from surrounding wastewater system (Lei et al., 2018). These transformations include physicochemical changes, transformations caused by agglomeration, aggregation, solubilisation, sedimentation, sorption, degradation, and deposition (Peijnenburg et al., 2015). A transformation may happen due to physical forces, chemical processes, and microbial degradation forces, including sunlight and different chemical substances (Peijnenburg et al., 2015). Moreover, the wastewater – sewage – sludge – agriculture – environmental exposure pathway has been identified for metal nanoparticles. The metal nanoparticles can be transferred into sulfides of nanoparticles within a very short time under the anaerobic conditions (Lombi et al., 2012). The fates of metal nanoparticles in the natural environments are still unclear, thus it demands safe disposal and proper management systems. Limited information is available in the existing literature on the management of used metal nanoparticles and nano-toxicology. The best approach is to recycle the nanoparticles for reuse multiple times. The used or exhausted nanoparticles can be trapped by use in the manufacturing of bricks. Otherwise, metal nanocomposites can be stored in a steel cylinder and buried deep in the earth. Recovered organic and inorganic contaminants also should be placed in cylinders and buries deep into the earth (Ahmad et al., 2016). Further research should be focus on the interaction among metal nanoparticles, laden contaminants, natural organic matter to ensure sustainable wastewater treatment.

4.3 Disposal and utilisation of magnetic biosorbents from wastewater

Safe disposal and management of magnetic biosorbents and adsorbed pollutants requires more research efforts (Mauter et al., 2018). The reuse options are illustrated in Figure 5. Separation of iron oxide/iron modified biosorbents is possible for reuse by magnetic sedimentation and centrifugal sedimentation (Matsuda et al., 2016). The recovery of magnetic biosorbents is a complicated and laborious process (Ahmad et al., 2016; Harikishore Kumar Reddy et al., 2017; Salt et al., 1995). The recycled magnetic biosorbents could be used in the construction industry to produce bricks and cement, while large quantities could affect the mechanical properties of the final products. Its use in agriculture for soil aggregation and retaining moisture may lead to accumulation of contaminants in soil, which can seriously affect the health of aquatic and human life (Ahmad et al., 2016). After the contaminants are separated, they can be disposed of in a controlled environment. Another effective option is to place the adsorbents in soil and use plants to phytoremediate these contaminants. These plants can then be used as raw materials for the synthesis of biochar.

5. Estimating the cost of magnetic biosorbents

The cost-effectiveness of adsorbents is usually overlooked but should be properly analysed to ensure the practicability of any adsorbents for large scale wastewater treatments. Most of the desired properties of adsorbents should be recovered through recycling to reduce operating costs. Magnetic biosorbents could be an efficient option due to easy separation by magnetic fields. However, the cost of magnetic adsorbents depends on the availability of raw biomass, cost of metal nanoparticles, and modification processes. Raw biomass is a cost-effective, globally accessible material for biochar production (Kaushal and Upadhyay, 2014; Mohan and Pittman Jr, 2006; Mohan et al., 2014c; Tran et al., 2015). The cost of biomass depends on the availability of raw materials, processing value, and market demand (Mohan and Pittman Jr, 2006). The cost of biochar production is much lower than the activated carbon, minerals, polymer, graphene, and CNTs (Guo et al., 2010; Jasper et al., 2010). On the other hand, metal or metal oxide nanoparticles are cheaper than carbon nanotubes, graphene, and other nanoparticles. Iron, nickel, and manganese oxide nanoparticles have magnetic characteristics that can help to separate and regenerate the saturated adsorbent materials and improve adsorption capacity (Ali et al., 2016; Bagheri and Julkapli, 2016; Pan et al., 2017; Yu et al., 2018). Re-cycling and reusing magnetic biosorbents more than once reduces the cost of wastewater treatment. The price of nanomaterials mainly depends on the pore size diameter. The larger the diameter, the lower the cost, and vice versa. Instead of using CNTs, carbon-based biomass materials can be employed because they have enormous potential for developing different functional groups on their surface at low or no cost. Consequently, biosorbents can serve as a viable alternative option instead of CNTs and graphene. At the same time, iron nanomaterials are one of the cost-effective options for remediation of wastewater (Abdul-Raheim et al., 2016). Nonetheless, more research is needed on the development of desired and low-cost magnetic nanoparticles so that they can be applied on an industrial scale. Minimising the production costs and developing or extending the limits of reusability will help considerably in solving the world's real water crisis.

6. Conclusion and future prospectives

Magnetic biosorbents have shown increasing potential in terms of their properties and efficiency in the remediation of organic and inorganic contaminants in wastewater. This review summarised the recent synthesis methods and modification procedures for magnetic biosorbents, their application in removal of a variety of contaminants, and recovery for recycling. Based on the review, the main conclusions are:

- Metal nanoparticles, especially iron, are the most cost-efficient, and advantageous modification materials due to their ferromagnetic properties for the preparation of magnetic biosorbents, which helps to improve the surface area, functional groups, active sites and consequently the removal efficiency. Nickel, copper, zinc, and zirconium based minerals and nanoparticles were also used for synthesis of metal induced biosorbents.
- Magnetic biosorbents can be synthesised by single step (pre-treatment), double step (post-treatment), mineral-supported, hydrothermal, microwave-assisted, and green synthesis methods. Conventional pyrolysis processes can develop various functionalities better than microwave-assisted biosorbents. The diverse functional groups are developed from the oxidation-reduction process during pyrolysis of the mixed metal or metal oxide nanoparticles or chemicals. Thus, the pre-treatment (single-step) modification method is considered as a simpler and highly efficient method due to the presence of maximum active sites. Minerals modified biosorbents have a comparatively low production cost than the conventional magnetic nanoparticles modified biosorbents. Hydrothermal modified biosorbents can reduce greenhouse gas emissions during synthesis of biosorbents but produce low surface area and porosity, which limits their application. The hydrothermal modified biosorbents are also considered as a single step modification. Recently, engineered magnetic biosorbents have attracted research attention because they are self-sustainable, and no artificial impregnation of metal nanoparticles is required.
- Various contaminants were evaluated for their removal by magnetic biosorbents in this review. Surface functional groups of MB contribute to the removal of ionic

contaminants through ion exchange, coordination, complexation and electrostatic interaction. Organic contaminants are generally adsorbed by hydrophobic interaction, interparticle diffusion, partition, and hydrogen bonding. However, non-persistent organic contaminants can be remediated through catalytic degradation by metal nanoparticles

- Magnetic biosorbents are one of the more efficient adsorbents in comparison with unmodified biochar, activated carbon, cellulose-based adsorbents, chemically modified plant waste, carbon-based nanomaterials (CNTs, Graphene), and minerals for removal of cations. Similarly, the highest adsorption capacity of anions was found in metal nanocomposites, and magnetic biosorbents in comparison with industrial waste materials, activated carbon, biochar, and polymers. Thus, magnetic biosorbents are efficient candidates for removal of cations and anions. Limited information has been published for removal of organic contaminants by magnetic biosorbents, which requires further research. However, magnetic biosorbents also seem to be a promising candidate for remediation of organic contaminants due to their high adsorption capacity and catalytic effect for degradation.

- It is demonstrated that sorption capacity of ionic contaminants will benefit from the development of different functional groups and surface charge of the magnetic biosorbents. Metals modified biosorbents can compete with cations adsorption if the net surface charges are negative. However, metal modification of biosorbents can enhance adsorption capacity of anionic contaminants as metal nanoparticles are not occupying positively charged sites of the magnetic biosorbents. In contrast, hydrophobicity, surface area, and porosity of adsorbents are predominant factors for sorption of organic contaminants.

- Magnetic biosorbents can be reused and recycled for remediation of contaminants up to 5-7 times without significantly losing their adsorption capacity at a lab-scale. Diluted acid, base and organic solvents including HCl, HNO_3, H_2SO_4, NaOH, methanol, and ethanol can serve for the separation of contaminants from magnetic biosorbents.

Further research can be conducted on the remediation of emerging organic contaminants, e.g., PFAS using magnetic biosorbents or their derivatives. Tailored or engineered magnetic biosorbents can be prepared to develop distinctive characteristics that are advantageous for the removal of target contaminants. Multiple functional groups on the surface of magnetic biosorbents and their chemistry favor higher adsorption capacity which does require further investigation. To date, only a few studies have examined the field application of magnetic biosorbents, which requires further effort for the potential commercialisation of magnetic biosorbents. Future research efforts on the development of a more efficient operating system for easy and fast separation of the adsorbents for real wastewater treatment plants are necessary. In such case, environmental scientists, economists, engineers, policy makers and governments should work together for optimised design, installation, and application of the technology.

Conflicts of interest

The authors declare that they do not have any conflicts of interest.

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Tables

Table 1: Preparation of magnetic biosorbents, their characteristics, sorption condition and performance for organic and inorganic contaminants removal.

Magnetic biosorbents (MB)	Preparation Methods	Solid: solution ratio (molarity of the solution)	Preparation conditions	Characteristics of synthesized MB	Background electrolytes	Sorption condition	Adsorption capacity (mg/g)	Isotherm and kinetics	Ref
Cottonwood and $AlCl_3 \cdot 6H_2O$ driven magnetic biosorbents	Conventional heating processes	- (5 M)	Pyrolyzed at 600 °C under N_2 flow for 1 h	-Rough and porous morphology, well-crystalline, and AlOOH flakes observed	-	t=12 h, D =0.1 g/50mg	15 mg/g for As(V)	Elovich model, L	(Zhang and Gao, 2013)
Magnetic biosorbents driven from cottonwoods	Conventional heating processes: single step	- (2.5M)	Pyrolysed the mixture at 600 °C under N_2 flow for 1 h	-Well crystalline, rough and porous surface -Highly ferromagnetic properties (69.2 emu/g)	-	T=24h D =0.1 g/50mg	31.5 mg/g for As (v)	L, PFO	(Zhang et al., 2013)
Hematite modified MB	Conventional heating processes	- (4.20 M)	Pyrolyzed the mixture at 600 °C under N_2 flow for 1 h	-High surface area, and strong magnetic properties	-	T=48 h D=2.5 g/L, pH=7, I.C=20 ppm	0.4 mg/g for As (v)	L, PSO	(Wang et al., 2015)
Co and Fe loaded MB	Conventional pyrolysis	1:4 (1 M)	Pyrolyzed at 950 °C for 2 h under N_2 flow for 1 h	Well crystallized	-	I. C=100 ppm, pH=8	99% adsorption for Cd^{2+}	PSO L	(Harikishore Kumar Reddy and Lee, 2014)
$FeCl_3$ load MB driven from pinewoods	Conventional pyrolysis (single step)	2 g of the sawdust in 20 ml 1.0 mol/L	Pyrolysis at 600 °C for 115 min under N_2 flow for 2 h.	-Exhibit spinel structure, smooth, porous surface	-	pH-8,	204.2 mg/g for As (v)	PSO L	(Liu et al., 2010)

		$FeCl_3$ solutio n		and highly magnetic.					
$Co(NO_3)_2$ derived MB	Microwave heating	1: 10 (2 mol/L $FeCl_3$ and 1 mol/L $Co(NO_3)_2$ mixed sol)	After sonication for 2 h, Power applied -640 W and frequency of MW was 2.45 GHz under nitrogen flow	-Well crystalline -High surface area (247 m^2/g) and porosity	-	pH=5.0, room temp, t=24 h	51. 7 mg/g for Cr(VI)	PSO, L	(Wang et al., 2013a)
$NiCl_2$ driven MB from bamboo.	Microwave heating	10 g of BC introduced in 1.0 M of $NiCl_2$	The mixtures put in an MW heating with 640 W and a frequency of 2.45 GHz for 6 min under a nitrogen flow	-High surface area and porosity -More smooth surface due to the presence of nickel on the surface	0.01 M, and 0.1 M $NaNO_3$	T = 273 °C	142.7 mg/g f or Ni^{2+}	PSO, L	(Wang et al., 2013b)
Iron loaded MB	Co-precipitation method	1: 2	Treated with 5 M NaOH solution at pH 10-11	Improved surface area, porosity	0.1 M $NaNO_3$	pH=5	89.0 mg/g for Cu^{2+}	PSO, L	(Han et al., 2015b)
ZNS-loaded MB	Calcination method	-	Calcinated at 180 °C for 0.5 h under a nitrogen flow	Well crystalline and superparamagneti c properties	-	T = 4h I.C = 150 ppm	367.65 mg/g for pb^{2+}	PFO, L	(Yan et al., 2015b)
Magnetic chitosan	Calcination method	-	Calcined at 200˚C for 16 h under a nitrogen flow	Well crystalline	-	pH 3-5, t = 0.3 h, I.C.= 100 ppm	120 mg/g for Cr(VI)	L, PSO	(Yu et al., 2013)
MnO_x-loaded Magnetic biosorbents	Conventional method: double step method	1:40, 1:10 and 3:5	Pyrolyzed at 600 °C under a nitrogen flow	Layered of MnOx were well dispersed on the biochar surface	0.01 mol/L $NaNO_3$	-	160 mg/g for Cu^{2+}	L, PSO	(Song et al., 2014)

Biochar/Mg-Al assembled nanocomposite	Conventional method	The solid/liquid ratio was fixed at 1:10	Pyrolyzed at 600 °C for 1 h under a nitrogen flow	-Highly crystalline structure	$MgCl_2$ as electrolyte	pH = 6, t = 3 h	887 mg g for P (highest ever reported)	PSO, L-F	(Jung et al., 2015)
Magnetic biosorbents	Novel one-step electro-magnetization.	-	Pyrolyzed at 600 °C for 1 h under a nitrogen flow	-Improve porosity and magnetic properties	-	-	382 mg/g for orange 7.	-	(Jung et al., 2016)
Magnetically modified biosorbents	Conventional method	-	Pyrolyzed at 600 °C for 2 h under a nitrogen flow	Strong magnetic properties	0.01-1 M of Na^+, K^+ NO_3^- SO_4^{2-}	pH = 4,	45.8 mg/ g for As (v)	L, PSO	(Zhou et al., 2017)
Chitosan modification of magnetic biosorbents	Conventional method	-	Pyrolyzed in a tube furnace at 600 °C under N_2 environment f or 1 h	Highly magnetized and high surface area	0.01 mol L^{-1} NaCl, $Ca(NO_3)_2$, $CaCl_2$, Na_3PO_4, Na_2SO_4 solution	pH = 2	120 mg/g for Cr (vi)	L, PSO	(Zhang et al., 2015)
Magnetic biosorbents	Conventional method	Mass of biochar and Fe_3O4 is 2:1	-Pyrolysis at 400 °C for 2 h under N_2 environment	High surface area, porosity and magnetic properties	-	pH = 8, t = 7h T = 323K, D = 1.0 g/L	197.96 mg/g for Cd^{2+}	L, PSO	(Zhou et al., 2018)
Magnetic biosorbents	Co-precipitation metho d	-	Heated to 80 °C and then 10 ml of ammonium hydroxide solutio n (28%) for 30 min	High sorption capacity	0.001−0.1 mol/ L $NaNO_3$	pH = 4-8	-	L, PSO	(Lalhmunsiam a et al., 2017)
Magnetic biosorbents	Conventional method	-	Pyrolyzed at 600 °C under N_2 flow rate	Well-developed crystal structure and high CEC	0.01 M $NaNO_3$	-	200-350 mg/g for Cd^{2+}	L, PSO	(Trakal et al., 2016)

(Note, L-Langmuir, F-Freundlich, PSO-pseudo second order, PFO-pseudo first order, t-time, T-temperature, D-dose, I.C-Initial concentration

Table 2: Major adsorption mechanisms and corresponding functional group.

Adsorption mechanism	**Removal**	**Corresponding functional group**
Electrostatic interaction	Ionic contaminants including heavy metal (loid) s and ionic organic contaminants	Carboxylic acid, Alkyne, Carboxyl, Alkane, Alkene, Fe–(O, OH), Mn-O, Cu-O, Zn-O.
Ion exchange	Heavy metal (loid) s and ionic organic compound	Hydroxyl, Carboxyl, Amino group, Carboxylic acid, Phenols, Fe-O/OH.
Chelation	Predominantly Heavy metal (loid) s	Carbonyl, Hydroxyl, carboxyl, C-S.
Complexation	Heavy metal (loid) s and organic contaminants	Thiol amino group, Amino group, Sulfonic group, Carboxylic acid, OH–, COOH-,Fe–O, Metal-O/OH.
Hydrophobic interaction	Predominantly organic contaminants	Carbonyl and aromatic structures.
Reduction	Predominantly less persistent organic contaminants including dyes	The metal-oxygen functional group (i.e., Fe-O, Mn-O, Ti-O, Al-O, Zr-O, Cu-O).

Data extracted from (Ahmad et al., 2014; Da'na, 2017; Gao et al., 2017; Nupearachchi et al., 2017; Oh et al., 2016; Singh et al., 2018; Tan et al., 2016; Thines et al., 2017; Yan et al., 2015a; Yang et al., 2019; Zeng et al., 2018; Zhou and Haynes, 2010; Zhu et al., 2017)

Table 3: The removal of organic contaminants by magnetic biosorbents

Adsorbents	Organic contaminants	Adsorption capacity (mg/g)	References
Magnetic biochar	4-nitrotoluene	100.0	(Saleh et al., 2016)
Magnetic biosorbents derived from coconut shell	Phenol	20.0	(Hao et al., 2018)
Magnetic biosorbents derived from pine sawdust	Sulfamethoxazole	13.8	(Reguyal et al., 2017)
Magnetic biochar	Nitrotoluene	80 %	(Chen et al., 2011)
Magnetic biochar prepared from ZVI	methylene blue (MB)	70 %	(Ahmed et al., 2018)
$FeCl_3$ loaded fruit waste	methylene blue (MB)	31.0	(Mubarak et al., 2015)
Magnetic biochar derived from corncob and Fe_3O_4	methylene blue (MB)	163.0	(Ma et al., 2015)
Magnetic biochar derived from microalgae	Tetracycline	95.0	(Peng et al., 2014)
Magnetic biochar prepared from coconut waste	Acid orange	404.0	(Zhang et al., 2007)
Magnetic biochar prepared from chicken bones	Rhodamine B dye	75%	(Oladipo and Ifebajo, 2018)
Magnetic biochar prepared from chicken bones	Tetracycline	75%	(Oladipo and Ifebajo, 2018)
Fe3O4 modified biosorbents	Crystal violet	278.55	(Sun et al., 2015)
AlOOH modified cotton wood-derived biochars	Methylene blue	85.0	(Zhang and Gao, 2013)
nZVI modified biochars from rice husk	Methylene blue	13 times higher than pristine biochar	(Devi and Saroha, 2014)
nZVI modified paper mill sludge	pentachlorophenol	Much higher than pristine biochar	(Han et al., 2015a)
Ferric chloride hexahydrate modified biosorbents	Methylene blue	265	(Mubarak et al., 2014)

Table 4: Desorption of magnetic biosorbents with solvents.

Adsorbent materials	Contaminants	Chemicals used	Desorption capacity (%)	No. of Recycling times	References
Magnetic biochar using Kans grass	As	NaOH (0.5 mol L^{-1})	70-90	-	(Baig et al., 2014)
Magnetic wheat straw	As	NaOH (0.1 mol L^{-1})	80.0	-	(Tian et al., 2011)
Fe_3O_4@organge peel	Cd	0.1 M HNO_3	98.5	5	(Gupta and Nayak, 2012)
Fe_3O_4@Chitosan	Cu	0.1 M EDTA	96.0	4	(Yuwei and Jianlong, 2011)
Fe_3O_4@Chitosan	Alizarin red	0.1 M NaOH	93.5	7	(Fan et al., 2012)
EDTA@Fe_3O_4@Sawdust	Cd	0.1 M HCl	99.0	5	(Kataria and Garg, 2018)
	Cd	0.1 M HNO_3	100.0	5	
	Cd	0.1 M H_2SO_4	100.0	5	
	Cd	0.1 M $Ca(NO_3)_2$	80.0	5	
	Cd	0.1 M $Na(NO_3)_2$	54.0	5	

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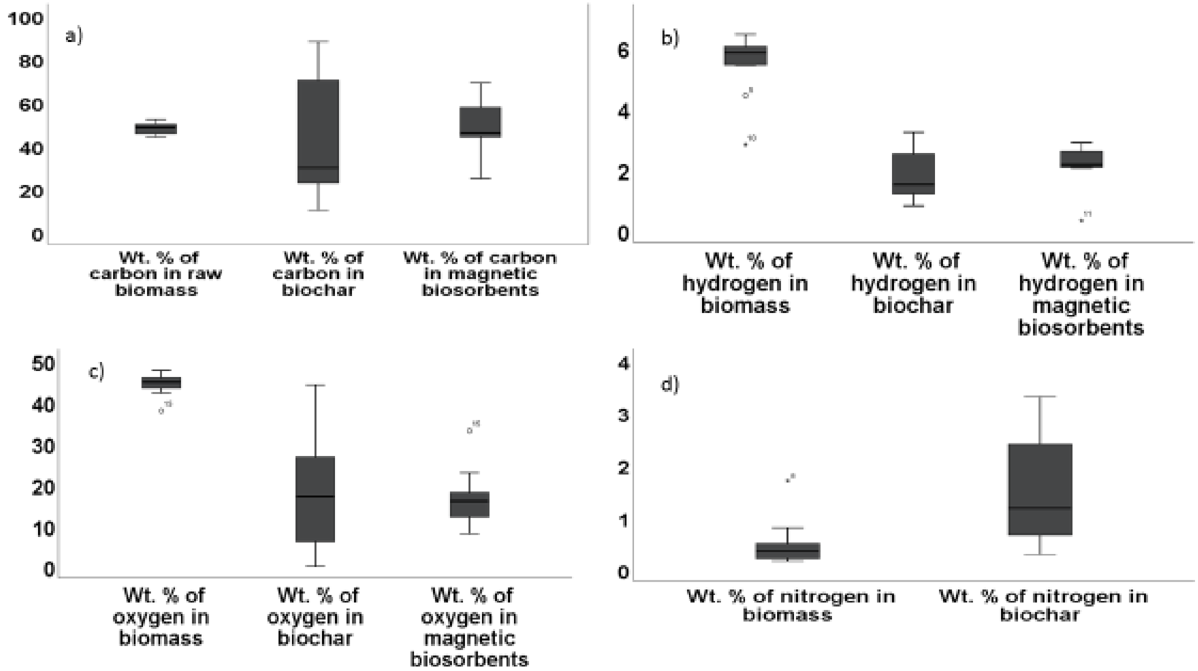

Figure 1: Variation of a) carbon b) hydrogen c) oxygen and d) nitrogen composition into biomass, biochar and magnetic biosorbents (Data extracted from (Ao et al., 2018; Bharathiraja et al., 2018; Chen et al., 2017; Liu et al., 2017; Moreira et al., 2017; Qian et al., 2015; Vassilev et al., 2013; Vassilev et al., 2012; Xu et al., 2016; Yang et al., 2007; Zhang et al., 2013; Zhang et al., 2019)).

Supplementary materials

Critical review of magnetic biosorbents: their preparation, application, and regeneration for wastewater treatment

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Table SM1. List of identified major functional groups in different magnetic carbonaceous materials.

Functional group / Adsorbents materials	C-C	C-O	C-H	C-N	O-H	N-H	N-N	CHO-	COO-	Fe-O	Others	Reference
Urban bio-waste derived magnetic adsorbent	✓	✓								✓		(Nisticò et al., 2018)
Magnetic pine biochar		✓								✓		(Reguyal et al., 2017)
Gum Kondagogu modified with magnetic nanoparticles	✓	✓	✓		✓	✓				✓		(Saravanan et al., 2012)
$CuFe_2O_4$ mixed activated carbon	✓			✓		✓	✓			✓		(Zhang et al., 2007)
Shellac coated iron oxide	✓				✓					✓		(Gong et al., 2012)
Calcium-based magnetic biochar		✓			✓					✓		(Wu et al., 2018)
Fe_3O_4@ layer double hydro oxide@ bio-Nano composite			✓		✓			✓	✓	✓		(Dinari and Tabatabaeian, 2018)
Novel magnetic biochar	✓	✓	✓			✓				✓		(Yap et al., 2017)
Fe_3O_4@sawdust Carbon		✓	✓		✓					✓		(Kataria and Garg, 2018a)
EDTA@ Fe_3O_4@sawdust Carbon		✓	✓		✓					✓		(Kataria and Garg, 2018a)
Pinewood-based magnetic biochar	✓	✓	✓		✓				✓	✓		(Pholosi et al., 2018)
Sugarcane bagasse derived iron modification	✓	✓		✓	✓					✓		(Montero et al., 2018)

Corncob husk modified with iron nanoparticles	✓	✓		✓	✓				✓		(Montero et al., 2018)
Iron impregnated Biochar	✓	✓	✓					✓	✓		(He et al., 2018)
Chitosan modified magnetic biochar		✓	✓		✓				✓	C-S	(Li et al., 2018)
Magnetic biochar composite		✓	✓		✓				✓	C-S	(Li et al., 2018)
Magnetic EDTA/chitosan/TiO_2			✓	✓	✓	✓		✓	✓	Ti-O	(Alizadeh et al., 2018)
Magnetic activated carbon	✓				✓				✓		(Han et al., 2015)
Magnetic nanoparticles with tea waste	✓		✓		✓				✓		(Lunge et al., 2014)
Magnetic oak bark biochar	✓	✓			✓			✓	✓		(Mohan et al., 2014b)
Magnetic oak wood biochar	✓	✓			✓			✓	✓		(Mohan et al., 2014b)
Bacterial cell nano stabilizer on sawdust with $MnFe_2O_4$		✓	✓			✓			✓		(Yan et al., 2016)
nZVI supported seed biochar materials	✓	✓	✓	✓	✓	✓		✓	✓	Ca-O, S-H, P-O	(Soleymanzadeh et al., 2015)
$MnFe_2O_4$ modified sawdust	✓	✓	✓	✓	✓	✓		✓	✓	Mn-O, Si-O	(Podder and Majumder, 2015)
Magnetic sludge-based biochar	✓	✓							✓		(Wang et al., 2017a)
Magnetic Litchi peel	✓	✓			✓	✓	✓	✓	✓		(Jiang et al., 2015)
Fe_3O_4-coated biochar	✓	✓		✓	✓			✓	✓		(Sun et al., 2015)
Saccharomyces cerevisiae and nanoparticles of iron oxide (Fe_3O_4)	✓			✓	✓				✓		(Zhu et al., 2018)
Magnetic biochar	✓					✓			✓		(Wang et al., 2018a)

Magnetic carbon composite	✓		✓	✓	✓		✓	(Wang et al., 2018a)
Fe_3O_4@C	✓	✓	✓	✓		✓	✓	(Tung et al., 2018)
Magnetic biosorbents from bagasse	✓	✓					✓	(Zahoor and Khan, 2018)

Note: here '✓' indicates the presence of a functional group

Table SM2: Removal of Cd ions by magnetic biosorbents and their physicochemical properties.

Adsorbents materials	Surface area (m^2/g)	Pore size (nm)	The volume of the pore (cm^3/g)	Wt. % of C	Wt. % of H	Wt. % of O	Wt. % of Fe	Adsorption capacity (mg/g)	References
Urban bio-waste derived magnetic biosorbents		80.00	-					50.00	(Nistico et al., 2018)
Iron modified rice straw (10%)	7.31	9.44	0.02	32.50	2.58	15.80	5.28	33.30	(Zhang et al., 2017)
Iron modified rice straw (5%)	26.45	7.35	0.04	43.80	2.79	15.90	4.87	14.90	(Zhang et al., 2017)
Iron modified rice straw (1% Fe)	23.36	10.84	0.05	46.10	2.86	16.20	1.97	24.70	(Zhang et al., 2017)
Rice straw biochar	3.20	45.01	0.01	49.30	2.46	17.90	0.79	34.20	(Zhang et al., 2017)
Magnetic rice straw biochar (10%)	5.70	26.02	0.02	44.10	2.08	13.80	8.25	35.20	(Zhang et al., 2017)
Magnetic rice straw biochar (5%)	5.99	24.51	0.02	44.20	2.10	12.00	8.12	38.70	(Zhang et al., 2017)
Magnetic rice straw biochar (1%)	14.62	18.62	0.04	45.50	2.06	11.20	3.03	33.90	(Zhang et al., 2017)
Gum Kondagogu modified with magnetic nanoparticles		15.00						106.80	(Saravanan et al., 2012)
Chitosan-based magnetic nanoparticles		25.00						9.21	(Liu et al., 2009)
Magnetic oak bark char	8.80		0.69	56.14	2.20			55.91	(Mohan et al., 2014a)
Magnetic oak wood char	6.10		0.48	69.02	0.32			10.13	(Mohan et al., 2014a)
Shellac coated iron oxide	56.91	20.00		24.86		22.74		18.80	(Gong et al., 2012)

Ferromanganese binary magnetic biochar	71.64	5.71	0.11	59.27	2.04	7.89	1.16	101.00	(Zhou et al., 2018)
$MnFe_2O_4$ biochar	30.28	4.15	0.03					181.49	(Wang et al., 2018b)
Coconut husk drove magnetic biochar				63.55		32.92	3.53	9.60	(Thines et al., 2017)
Fe_3O_4@ layer double hydro oxide@ bio-nano composite	178.70	40.00	-					258.00	(Dinari and Tabatabaeian, 2018)
Novel magnetic biochar	834.00							4.71	(Yap et al., 2017)
Fe_3O_4@sawdust carbon	30.15	3.20	0.15					51.00	(Kataria and Garg, 2018a)
EDTA@ Fe_3O_4@sawdust carbon	14.00	3.18	0.09					63.00	(Kataria and Garg, 2018a)
Chitosan modified magnetic biochar	27.62	0.12		50.83	3.93		1.33	137.30	(Li et al., 2018)
Magnetic biochar composite	112.33	0.09		60.63	1.81		2.83		(Li et al., 2018)
Magnetic EDTA/chitosan/TiO_2		40.00						209.21	(Alizadeh et al., 2018)
Magnetic oak bark biochar	8.80		0.69	56.14	2.19			99.73	(Mohan et al., 2014b)
Magnetic oak wood biochar	6.10		0.48	69.02	2.77			66.22	(Mohan et al., 2014b)
Magnetic peel drove iron oxide				68.44		18.38	8.38	45.66	(Ruthiraan et al., 2017)
Fe_3O_4 coated Sawdust carbon		100.00		53.70		39.50	6.30	51.00	(Kataria and Garg, 2018b)

EDTA@Fe_3O_4@sawdsut				63.40		30.70	5.40	63.30	(Kataria and Garg, 2018b)
Magnetic biochar	890.00	2.28	0.68					62.50	(Ruthiraan et al., 2015)
Magnetic oak wood	6.10		0.48	69.02	2.70			7.40	(Mohan et al., 2014b)
Magnetic oak bark	8.80		0.69	56.14	2.19			2.87	(Mohan et al., 2014b)

Table SM3: Removal of arsenic by magnetic biosorbents and their physicochemical properties.

Adsorbents materials	Surface area (m^2/g)	Pore size (nm)	Volume (cm^3/g)	Wt. % of C	Wt. % of H	Wt. % of O	Wt. % of Fe	Adsorption capacity (mg/g)	References
Iron modified rice straw (10%)	7.31	9.44	0.02	32.50	2.58	15.80	5.28	46.30	(Zhang et al., 2017)
Iron modified rice straw (5%)	26.45	7.35	0.04	43.80	2.79	15.90	4.87	8.70	(Zhang et al., 2017)
Iron modified rice straw (1 % Fe)	23.36	10.84	0.05	46.10	2.86	16.20	1.97	9.50	(Zhang et al., 2017)
Rice straw biochar	3.20	45.01	0.01	49.30	2.46	17.90	.79	10.20	(Zhang et al., 2017)
Magnetic rice straw biochar (10%)	5.70	26.02	0.02	44.10	2.08	13.80	8.25	40.80	(Zhang et al., 2017)
Magnetic rice straw biochar (5%)	5.99	24.51	0.02	44.20	2.10	12.00	8.12	39.30	(Zhang et al., 2017)
Magnetic rice straw biochar (1%)	14.62	18.62	0.04	45.50	2.06	11.20	3.03	67.40	(Zhang et al., 2017)
Magnetic biochar using Kans grass (double step)	38.22	18.57	0.15	51.50		39.16	9.34	1.64	(Baig et al., 2014)
Magnetic biochar using Kans grass (double step)	38.22	18.57	0.15	51.50		39.16	9.34	1.30	(Baig et al., 2014)
Magnetic biochar using Kans grass (double step)	31.16	16.37	0.10	61.11		30.58	8.31	1.75	(Baig et al., 2014)
Magnetic biochar using Kans grass (double step)	31.16	16.37	0.10	61.11		30.58	8.31	1.41	(Baig et al., 2014)

Magnetic biochar using Kans grass (single step)	27.60	14.80	0.11	58.57		26.75	14.68	1.71	(Baig et al., 2014)
Magnetic biochar using Kans grass (single step)	27.60	14.80	0.11	58.57		26.75	14.68	1.92	(Baig et al., 2014)
Magnetic biochar using Kans grass (single step)	31.45	21.96	0.17	54.39		27.37	18.24	2.04	(Baig et al., 2014)
Magnetic biochar (single step)	31.45	21.96	0.17	54.39		27.37	18.24	3.13	(Baig et al., 2014)
Pinewood-based magnetic biochar	54.80	10.17	0.15					13.86	(Pholosi et al., 2018)
Sugarcane bagasse derived iron modification				20.62		25.06	54.32	20.00	(Montero et al., 2018)
Corncob husk modified with iron nanoparticles				21.98		21.97	52.08	50.00	(Montero et al., 2018)
Iron impregnated biochar	293.13	5.80	0.10	59.95	1.75	18.08	6.05	6.80	(He et al., 2018)
Magnetic nanoparticles with tea waste		25.00						189.69	(Lunge et al., 2014)
nZVI with pine driven biochar	211.70			72.16			10.50	124.50	(Wang et al., 2017b)
Hematite modified biochar	193.10			51.70	1.40	43.10	2.95	.42	(Wang et al., 2015c)
Sawdust modified with MnFe2O4	22.98		0.01					506.98	(Podder and Majumder, 2015)
Magnetically coated wood biochar								3.14	(Zhang et al., 2013)
Pinewood magnetic biochar	193.10			51.70	1.40	43.10	2.95	.43	(Wang et al., 2015c)

Magnetic pine sawdust	349.00						39.00	204.20	(Liu et al., 2010)
Magnetic Loblolly pinewood	463.10		0.02	78.90	1.86	14.58		.59	(Wang et al., 2015a)
Birnessite impregnated pinewood	67.40		0.07	61.54	1.85	27.65		.91	(Wang et al., 2015a)
Microwave-assisted magnetic biochar from wheat straw	13.00		0.02	26.40	1.00		40.40	25.60	(Zubrik et al., 2018)
Conventional heating magnetic biochar from straw	119.30		0.01				35.50	17.89	(Zubrik et al., 2018)
Manganese oxide with biochar	463.10		0.02	78.95	1.86	14.58		.59	(Wang et al., 2015a)
Birnessite with biochar	67.40		0.07	61.54	1.85	27.65		.91	(Wang et al., 2015a)
Magnetic modification of biochar with hematite	193.10			51.70	1.40	4.31	2.95	.40	(Wang et al., 2015d)
Manganosite modified pine biochar	463.10		0.02	78.95	1.86	14.58	4.19	.59	(Wang et al., 2015b)
Birnessite modified biochar	67.40		0.07	61.54	.25	27.65	8.14	.91	(Wang et al., 2015b)
Iron oxide (ferrous) loaded AC	987.00	5.88	0.43					2.02	(Tuna et al., 2013)
Iron oxide (ferric) loaded AC	1231.00	5.88	0.57					3.01	(Tuna et al., 2013)
Magnetic activated carbon with $ZnCl_2$	806.00		0.25	76.80		21.70		4.56	(Nieto-Delgado and Rangel-Mendez, 2012)
Magnetic activated carbon with H_3PO_4	1058.00		0.48	69.10		24.60		4.56	(Nieto-Delgado and Rangel-Mendez, 2012)
Iron impregnated biochar				79.09		16.75	3.03	2.16	(Zhu et al., 2014)
Magnetic biochar from peanut	18.20			46.24		31.17		33.70	(Pan et al., 2015)

straw					
Magnetic biochar from rice straw	11.30	39.69	41.98	27.00	(Pan et al., 2015)

Table SM4: Correlation coefficients between the physicochemical parameter of magnetic biosorbents (MB) and adsorption capacity.

Physicochemical factor of MB / Adsorption capacity (mg/g)	Surface area (m^2/g)	pore size (nm)	Wt. % of H	Wt. % of O	Wt. % of Fe
Cadmium (Cd)	-.023	.006	**.418**	-.167	-.284
	(22)	(20)	**(15)**	(13)	(14)
Arsenic (As)	-.116	**.248**	**.295**	**-.295**	**.339**
	(37)	**(20)**	**(18)**	**(32)**	**(28)**

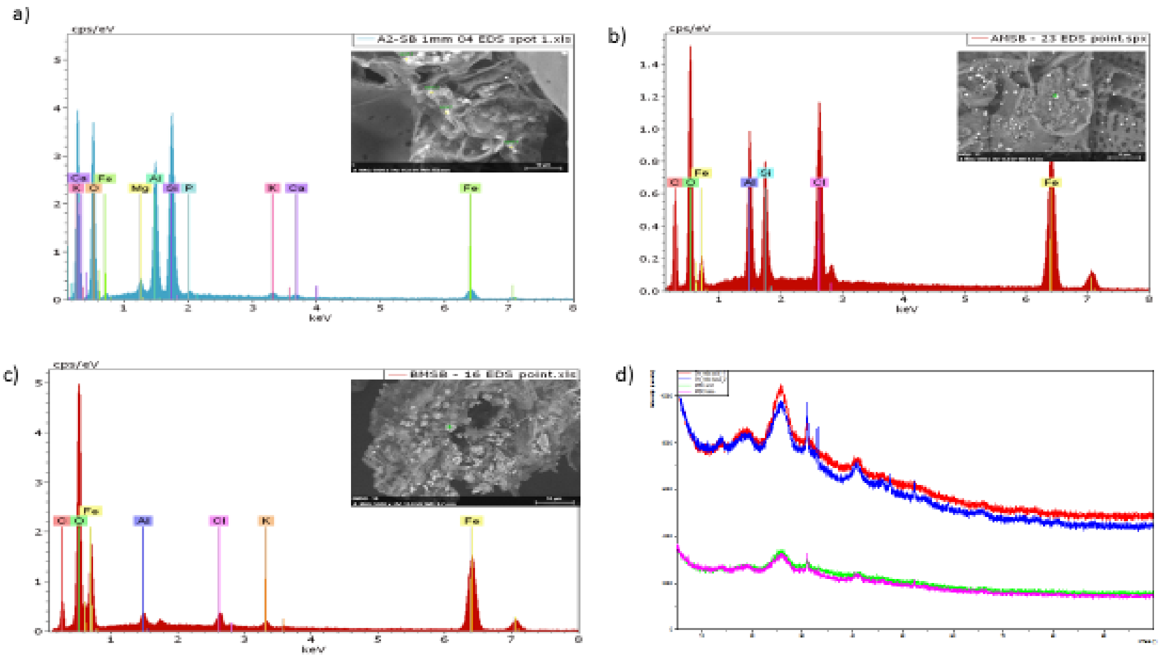

Figure SM1. SEM micrograph and EDS spectra of (a) unmodified sugarcane bagasse (SB); (b) acid-modified magnetic biosorbents from SB; (c) base modified magnetic biosorbents from SB and (d) XRD spectra of raw biosorbents and metal-doped biosorbents.

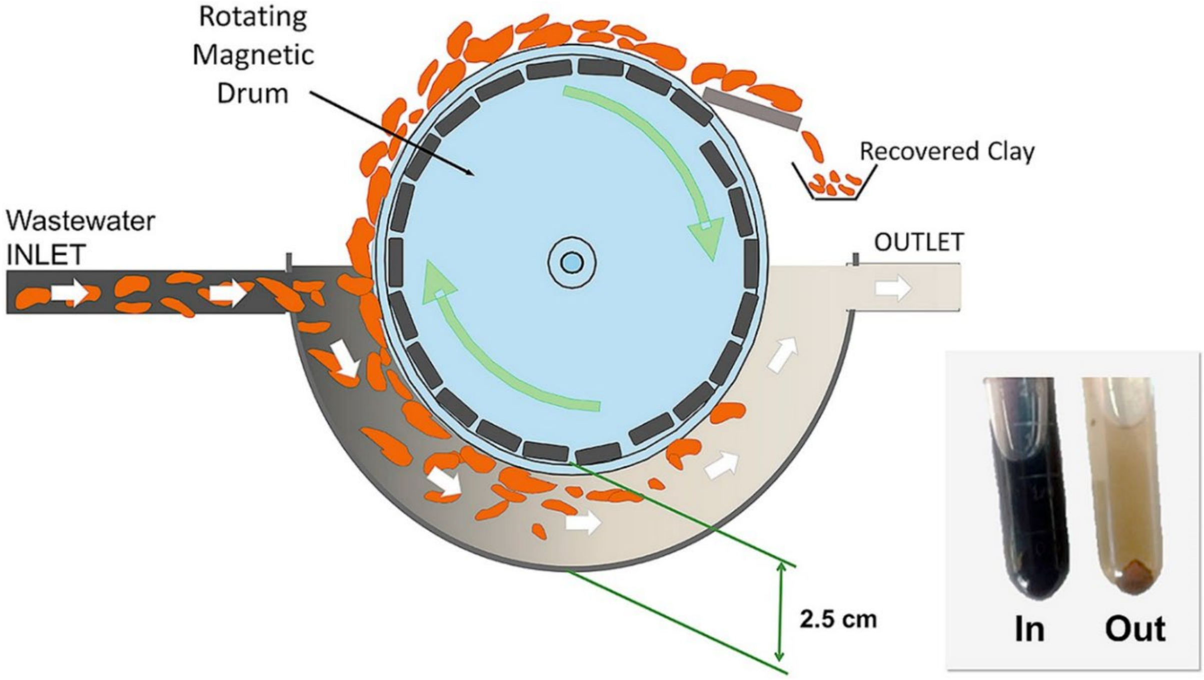

Figure SM2: Operation diagram of the magnetic separator for application and recovery of magnetic adsorbents (derived from clay minerals) for pilot scale application (Salinas et al., 2018).

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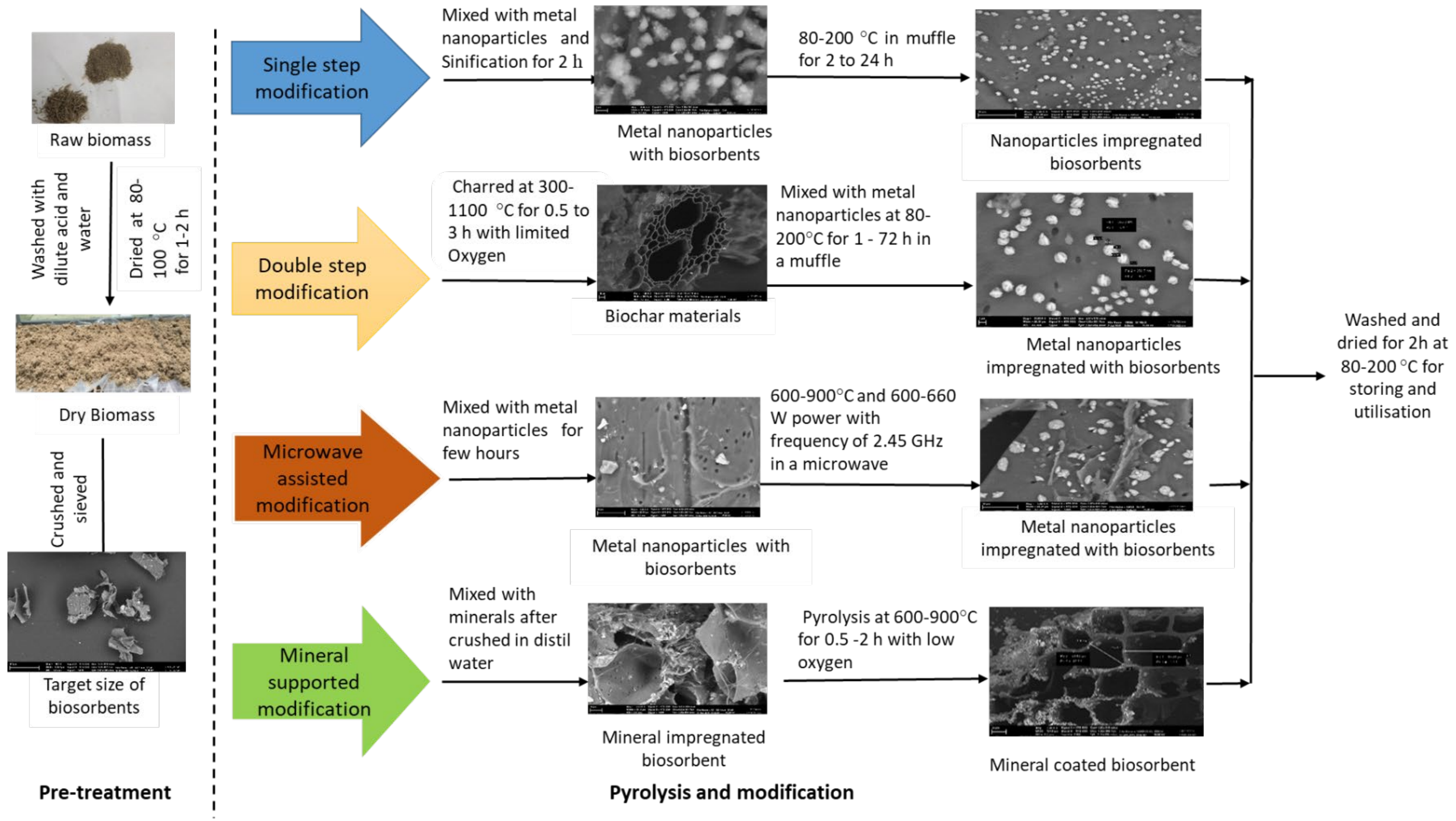

Figure 2: The synthesis of magnetic biosorbents using different methods: single-step modification, double step modification, microwave-assisted modification, and minerals supported modification.

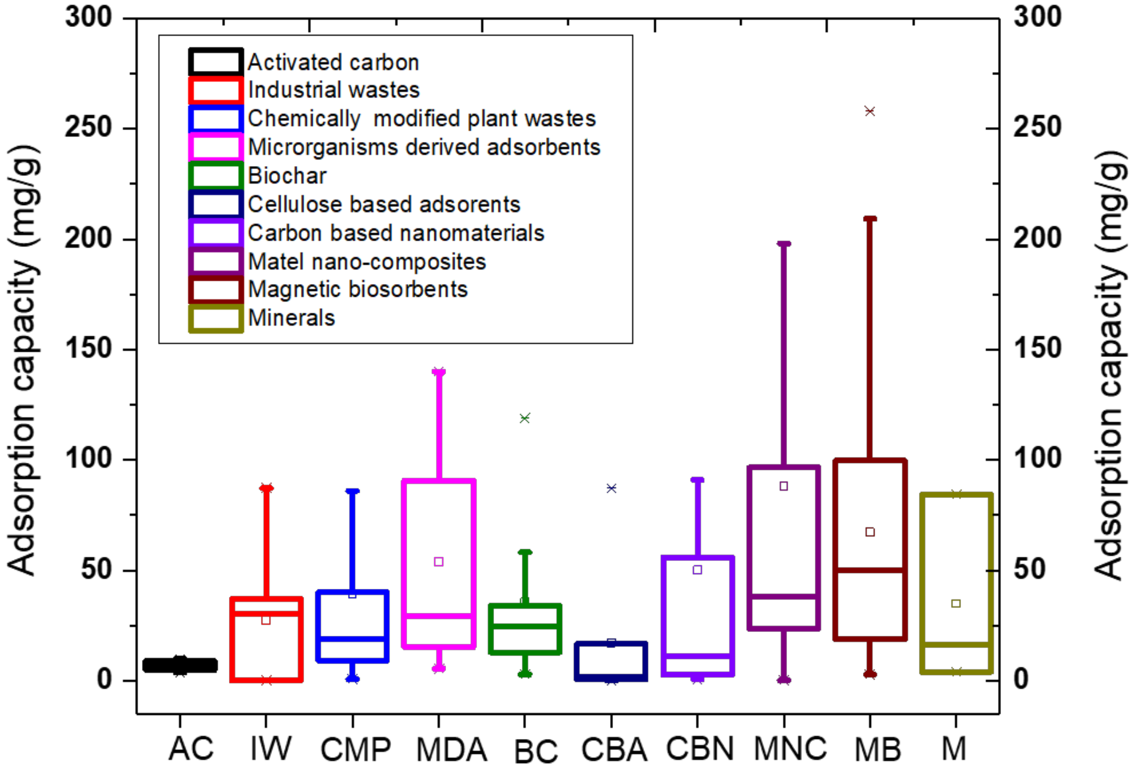

Figure 3: Comparison of the adsorption capacity of Cd by a wide range of sorbents (Data extracted from (Adusei-Gyamfi et al., 2019; Babel and Kurniawan, 2003; Bailey et al., 1999; Burakov et al., 2018; Dai et al., 2018; Deshpande, 2017; Joseph et al., 2019; Kumar et al., 2015; Naghizadeh, 2015; Pal et al., 2017; Pyrzynska, 2019; Sen Gupta and Bhattacharyya, 2011; Sizmur et al., 2017; Sud et al., 2008; Vikrant et al., 2019; Wan Ngah and Hanafiah, 2008; Xu et al., 2018; Yadanaparthi et al., 2009). AC-activated carbon; IW-industrial wastes; CMP-chemically modified plant wastes; MDA-microorganisms derived adsorbents; BC-biochar; CBA-cellulose based adsorbents; CBN-carbon based nanomaterials; MNC-metal nano-composite; MB-magnetic biosorbents; M-minerals.

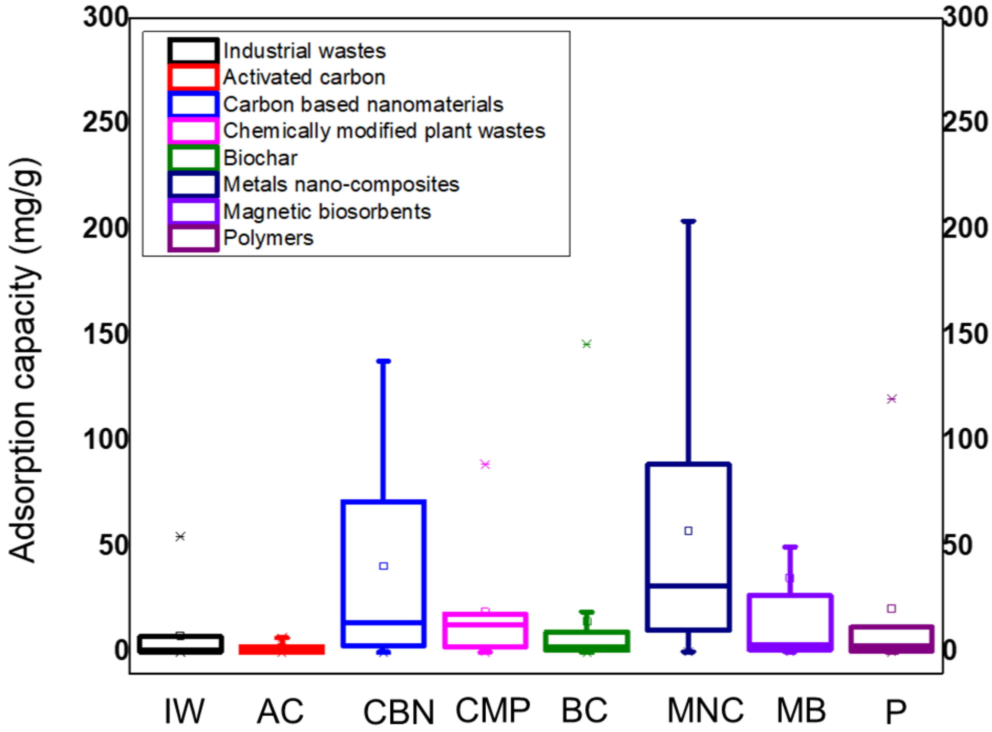

Figure 4. The removal capacity of Arsenic (III and V) by a wide range of adsorbents (Data extracted from (Bhatnagar et al., 2011; Deliyanni et al., 2015; Ihsanullah et al., 2016; Jain et al., 2016; Khin et al., 2012; Kyzas et al., 2014; Mohan and Pittman Jr, 2007; Ng et al., 2004; Nguyen et al., 2013; Rosales et al., 2017; Sharma et al., 2009; Siddiqui et al., 2019; Siyal et al., 2018; Xu et al., 2012; Zare et al., 2018; Zhou and Haynes, 2010). IW-industrial wastes; AC-activated carbon; CBN-carbon based nanomaterials; CMP-chemically modified plant wastes;BC-biochar; MNC-metal nano-composite; MB-magnetic biosorbents; P-polymer

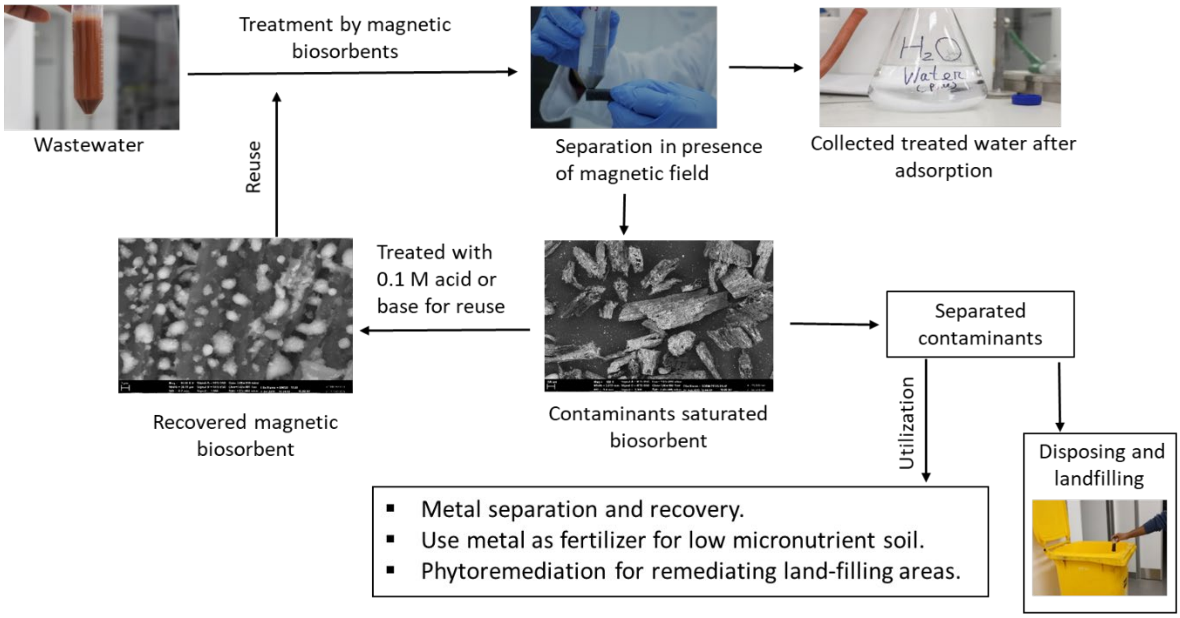

Figure 5: Application and regeneration processes for magnetic biosorbents.